

1 Strain relaxation and multidentate anchoring in n-type perovskite transistors and logic circuits

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35 **The engineering of tin halide perovskites has led to the development of p-type transistors with**
36 **field-effect mobilities of over 70 cm² V⁻¹ s⁻¹. However, due to their background hole doping,**
37 **these perovskites are not suitable for n-type transistors. Ambipolar lead halide perovskites are**
38 **potential candidates, but their defective nature limits electron mobilities to around 3-4 cm² V⁻¹**
39 **s⁻¹, which makes the development all-perovskite logical circuits challenging. Here, we report**
40 **formamidinium lead iodide perovskite n-type transistors with field-effect mobilities of up to 33**
41 **cm² V⁻¹ s⁻¹ measured in continuous bias mode. This is achieved through strain relaxation of the**
42 **perovskite lattice using a methyl ammonium chloride additive, followed by suppression of**
43 **undercoordinated lead through tetramethyl ammonium fluoride multidentate anchoring. Our**
44 **approach stabilizes the alpha phase, balances strain, and improves surface morphology,**
45 **crystallinity, and orientation. It also enables low-defect perovskite–dielectric interfaces. We use**
46 **the transistors to fabricate unipolar inverters and eleven-stage ring oscillators.**

47

48 **Main**

49 Metal halide perovskites are a class of semiconductors suitable for use in cost-effective thin-film
50 transistors (TFTs)¹⁻¹³. Recent efforts to optimize tin (Sn) halide perovskites using halide anion
51 engineering^{14,15}, morphology and crystallinity manipulation¹⁶, and A-site cation engineering¹⁷, have
52 led to p-type transistors with field-effect hole mobilities (μ_h) of over 70 cm² V⁻¹ s⁻¹. However,
53 perovskite channels based on tin are not suitable for n-type TFTs due to their high background hole
54 doping^{18,19}. Lead halide perovskites have ambipolar charge transport properties¹³, and are thus more
55 suitable for n-type transistors. However, ionic defects, especially undercoordinated lead present at
56 the film surface and grain boundaries, act as charge carrier traps limiting electron mobilities (μ_e) to
57 3-4 cm² V⁻¹ s⁻¹. They also screen the applied gate potential causing severe hysteresis. Therefore, most
58 perovskite TFTs are measured in an electrically pulsed mode with prior poling¹⁸. These defective
59 sites also initiate perovskite hydrolysis further compromising the performance and stability of the
60 fabricated devices²¹⁻²³. To build complementary logical circuits, the development of n-type
61 perovskite TFTs with performance comparable to their p-counterparts is required.

62 The cubic α -phase of formamidinium lead triiodide (α -FAPbI₃) has been shown to be a
63 promising perovskite semiconductor for photovoltaics^{24,25}. With a low electron effective mass of
64 ~ 0.2 , the perovskite is also suitable for n-type TFTs²⁶. However, due to the slightly overlarge ionic
65 radius of FA⁺ cation, FAPbI₃ exhibits a strained lattice at room temperature that drives phase
66 transformation into a defective delta (δ)-phase²⁴. Several strategies that use doping or additive

67 engineering, including the use of a methylammonium chloride (MACl) additive^{24,25}, have led to
68 successful photovoltaic performance of FAPbI₃²⁵⁻²⁷.

69 In this Article, we show that a MACl additive approach can be used to regulate strain and
70 stabilize the α -phase of FAPbI₃, while also improving the perovskite film morphology and
71 crystallinity/orientation. We also find that the critical perovskite–dielectric interface still has a large
72 amount of undercoordinated lead cations, resulting in poor transistor performance. To overcome this,
73 we use tetramethylammonium hexafluorophosphate (TMA-PF₆) multidentate anchoring that decreases
74 undesirable trap density at the surface, bulk and the perovskite–dielectric interface. The resulting
75 perovskite can be used as a n-channel material for top-contact bottom-gate (TCBG) TFTs using
76 molybdenum (Mo) gate electrodes. The transistors exhibit representative electron mobilities of
77 25.15 cm² V⁻¹ s⁻¹ (up to 33 cm² V⁻¹ s⁻¹ upon further optimization) and 18 cm² V⁻¹ s⁻¹ when
78 embedding hafnium dioxide (HfO₂) and silicon dioxide (SiO₂) gate dielectrics, respectively
79 (measured in continuous mode, at room temperature in the dark)²⁸. The devices also exhibit
80 negligible hysteresis and high operational stability under negative and positive bias stress for
81 150,000 seconds. We use our optimized transistors to fabricate all-perovskite unipolar inverters and
82 11-stage ring oscillators

83

84 **Strain relaxation**

85 To suppress lattice strain and stabilize the α -phase of FAPbI₃, we inserted MACl additive into the
86 perovskite precursor in an optimized molar ratio of 35%. The precursor was next diluted with
87 tetrahydrofuran (THF) to adjust the channel thickness to around 40-50 nm (Supplementary Fig. 1a-d).
88 MACl additive substantially improves the film morphology as evidenced by the top-view scanning
89 electron microscopy (SEM) images shown in Fig. 1a-d and Supplementary Fig.1e. Films spin-coated
90 from 35% MACl (termed hereafter as α -FAPbI₃) added solutions exhibit the best morphology. THF
91 dilution and TMA-PF₆ surface treatment preserve the improved film morphology. Moreover, MACl-
92 additive induces better crystallinity (which is again preserved upon THF dilution and TMA-PF₆
93 surface treatment) (Fig. 1b and Supplementary Fig. 1f). The X-ray diffraction (XRD) patterns of all
94 MACl-added films exhibit two dominant peaks at around 14° and 28°, which are assigned to (001)
95 and (002) crystal planes of the α -FAPbI₃ phase²⁹. On the contrary, pristine FAPbI₃ is dominated by
96 the PbI₂ peak at 12.5° (which is a degradation by-product induced by perovskite hydrolysis³⁰) and the
97 δ -phase appeared at 11.6°. A slight shift of 0.01° in the (001) plane peak position upon the addition
98 of MACl is observed, which indicates a small reduction to inter-planar spacing (from 0.632274 to
99 0.631826 nm, Supplementary Table 1). This can be due to partial replacement of FA⁺ (ionic radius
100 0.19-0.22 nm) with the smaller MA⁺ (0.18 nm)³¹, in accordance to previous reports³². The inter-

101 planar spacing further decreases with THF dilution (0.63138 nm) and TMA-PF₆ treatment (0.63093
102 nm). MACl additive also increases the perovskite crystallite size (from around 41 nm to 74 nm,
103 Supplementary Table 1), which is further increased (to 80 nm) upon dilution with THF (in an
104 optimized ratio of 1:3 %v/v to adjust the perovskite thickness to 50 nm). These results show that
105 MACl addition suppresses the perovskite transformation into the yellow δ -phase while also alleviates
106 perovskite hydrolysis to PbI₂.

107 In addition to stabilizing α -FAPbI₃ phase, crystal orientation is also vital as it dictates charge
108 transport within the perovskite channel. We carried out grazing-incidence wide-angle X-ray
109 scattering (GIWAXS) measurements to determine the crystal orientations of the four different types
110 of FAPbI₃ perovskite films. Representative results of the 2D GIWAXs patterns are shown in Fig. 1f-i,
111 whereas the corresponding in-plane (IP) and out-of-plane (OOP) 1D line-cut profiles are shown in
112 Supplementary Fig. 2. The pattern of the pristine FAPbI₃ film exhibits apparent diffraction rings (Fig.
113 1f), a strong indication that the crystal structure is randomly orientated within the film. In stark
114 contrast, with the addition of MACl in the perovskite precursor the prepared films (without and with
115 THF dilution) reveal discrete and sharp Bragg spots (Fig. 1g,h), suggesting a high degree of vertical
116 orientation of the inorganic frameworks with respect to the substrate. A similar GIWAXS pattern is
117 obtained in the case of TMA-PF₆ treated film, indicating that the perovskite film morphology,
118 crystallinity and orientation are mainly benefited by the addition of MACl. These improvements are
119 consistent with the enhancement in steady-state photoluminescence (PL) and longer carrier lifetime
120 estimated by the transient PL (TRPL) decay curves of different FAPbI₃ films (Supplementary Fig. 3
121 and Supplementary Table 2). Notably, TMA-PF₆ post-treatment further enhances the PL signal and
122 carrier lifetime. The TRPL decay curves of such films show distinct mono-exponential decay, which
123 is indicative of effective defect passivation³³.

124 To explore the interactions between different species (i.e., precursors, additive and surface
125 modifier) in the different perovskite formulations, we first conducted nuclear magnetic resonance
126 (NMR) experiments in liquid phases where freely moving molecules closely approach one another.
127 Proton NMR (¹H-NMR) spectra of FAI in DMSO-d₆ show two sharp closely packed peaks at ~8.88
128 ppm originating from the FA's amine group³⁴ (Fig. 2a,b and Supplementary Fig. 4). Upon MACl
129 addition, these sharp peaks disappear and a broad peak appears at 8.6 ppm, which progressively
130 shifts towards higher fields (lower chemical shift) upon gradual addition of MACl up to a molar ratio
131 of 1:2. We ascribe this peak to the overlap of the resonance signals from NH₂, NH₂⁺ (FAI), and NH₃⁺
132 (MACl), shifting towards higher and lower fields, respectively³⁵. This suggests that the electron
133 density around the hydrogen atoms of MA⁺ decreases (whereas electron density around FA⁺
134 increases) in the FAI:MACl blend, indicating hydrogen-bonding between MA and FA cations.

135 Besides their up-field shift, NH_2 and NH_2^+ proton peaks significantly decrease in intensity which
136 suggests partial substitution of FA^+ sites with MA^+ . Fourier transform infrared (FTIR) spectra of
137 $\text{FAI}:\text{MACl}$ and $\text{FAPbI}_3:\text{MACl}$ films show shifts of the N-H vibration bands of FA^+ (i.e., the $\delta(\text{NH}_2)$
138 band at $\sim 1700\text{ cm}^{-1}$) towards lower wavenumber (Supplementary Fig. 5), indicating coordination
139 bonding (i.e., hydrogen bonding) between FA^+ and MA^+ species. However, the resonance signal of
140 NH_3^+ in MA cation is shifted towards the lower field after interacting with PbI_2 , indicating an
141 increase in electron density (deshielding effect). This can be due to interaction of MA^+ with I^- , which
142 is less electronegative compared to Cl^- . Strong interaction between PbI_2 and MACl is also concluded
143 by the suppression of most of the FTIR modes in the mixed film. These NMR and FTIR studies
144 suggest partial substitution of FA^+ sites with MA^+ and extended coordination (i.e., hydrogen bonding)
145 between FA^+ and MA^+ present in the perovskite lattice. Additionally, the resonance signals of F^- in
146 ^{19}F NMR and of protons in ^1H NMR of TMA-PF_6 exhibit pronounced shifts upon the interaction
147 with FAI , PbI_2 and MACl , indicating extended hydrogen (i.e., $\text{TMA}^+\dots\text{FA}^+$, $\text{F}^-\dots\text{FA}^+$, $\text{TMA}^+\dots\text{MA}^+$
148 and $\text{F}^-\dots\text{MA}^+$) and halogen ($\text{F}^-\dots\text{I}$) bonding coordination (Fig. 2c,d and Supplementary Figs 6,7). The
149 significant suppression of the FTIR spectrum of $\text{PbI}_2:\text{TMA-PF}_6$ blend film is indicative of strong
150 Lewis acid-base interaction between Pb and F ionic species (Supplementary Fig. 8).

151

152 **Structural defect passivation**

153 To further explore the benefits of MACl additive and TMA-PF_6 treatment, we performed X-ray and
154 ultra-violet photoelectron spectroscopy (XPS/UPS) studies on pristine, MACl -added (without and
155 with THF dilution) and TMA-PF_6 treated FAPbI_3 films. Figure 2e shows the Pb 4f XPS peaks of the
156 different FAPbI_3 films. Regarding the pristine perovskite, a large amount of undercoordinated $\text{Pb}^{+\delta}$
157 can be concluded from the large shift of the core level peaks towards lower binding energies (BEs)
158 (the oxidation state δ of such species can be $\delta < +2$ as they do not bond with electronegative halides)
159 ³⁶. When adding MACl into the precursor, the Pb 4f peaks shift toward higher BEs, which
160 correspond to +2 oxidation state. This shift can be due to their coordination with electronegative ions
161 (such as Cl^-) through Lewis acid-base ($\text{Pb}^{2+}-\text{Cl}^-$) interactions. It implies more well-coordinated Pb
162 sublattices and a lower density of structural imperfections (i.e., grain boundaries) in the $\alpha\text{-FAPbI}_3$.
163 Similarly, I 3d core level peaks of MACl -added films are shifted towards higher BEs (i.e., higher
164 oxidation state) compared to pristine one (Fig. 2f). In the ideal perovskite lattice, the theoretical I/Pb
165 stoichiometric value should be close to 3, whereas in PbI_2 is close to 2³⁷. The increase in oxidation
166 state of I^- in $\alpha\text{-FAPbI}_3$ indicates reduced PbI_2 formation. Notably, Pb 4f and I 3d depth XPS
167 measurements taken in pristine FAPbI_3 (Supplementary Fig. 9) show very small shifts towards
168 higher BEs when move away from the perovskite surface. This is indicative of the presence of

169 significant amount of undercoordinated $\text{Pb}^{\delta+}$ inside the bulk material (besides the surface), highly
170 compromising the TFT performance. MACl addition suppresses the amount of all these species,
171 which is in agreement to a small work-function (W_F) increase for the optimized T-FAPbI₃ sample³⁸
172 (Supplementary Fig. 10).

173 However, Pb 4f depth XPS measurements taken in the MACl-added samples (Supplementary
174 Fig. 11a) show a significant shift towards lower BEs ($+\delta$ oxidation state) an indication for remaining
175 undercoordinated Pb in the bottom of the bulk perovskite. The origin for such XPS peaks shift is not
176 well understood but it could imply that MACl additive spontaneously enriches the top side of the
177 perovskite film³⁹. This is further corroborated by the decrease in the intensity of the Cl 2p XPS signal
178 of the same film when move from the surface towards the bottom (Supplementary Fig. 11b). As a
179 result, detrimental undercoordinated Pb is still present at the perovskite/dielectric interface, hence
180 negatively affecting the TFT performance. When treating the α -FAPbI₃ film with TMA-PF₆, both Pb
181 4f and I 3d XPS peaks shift towards lower BEs, indicating an interaction with electronegative species
182 (such as F⁻). Notably, depth XPS measurements reveal that F anions are nearly homogeneously
183 incorporated into the bulk and surface of the perovskite material (Supplementary Fig. 11c). No sign
184 of F⁻ was found in the pristine perovskite (Supplementary Fig. 9b, inset). Moreover, I 3d and Pb 4f
185 depth XPS peaks do not shift from the surface towards the film bottom; an indication of
186 homogeneous composition throughout the bulk and surface (Supplementary Fig. 11d and inset). Our
187 findings, which suggest strain relaxation via MA⁺ substitution and multidentate anchoring through
188 hydrogen, halogen and Lewis acid-base interactions, are illustrated in Fig. 2g and Supplementary Fig.
189 11e. Notably, density functional theory (DFT) simulations strongly support these findings. They
190 indicate that TMA-PF₆ is coordinated to the perovskite bulk and surface through an extended
191 network of halogen (F⁻...I⁺ and F⁻...Cl⁺) and hydrogen (between F⁻ and H of polar NH₂, NH₂⁺ (FAI)
192 and NH₃⁺ (MACl)) bonds and also Pb²⁺...F⁻ bonds⁴⁰⁻⁴² (Fig. 2h,i, Supplementary note 1,
193 Supplementary Figs 12-14 and Supplementary Tables 3-6).

194

195 **Performance of n-channel α -FAPbI₃ transistors**

196 We next applied our optimized T-FAPbI₃ perovskite as the channel material in transistors with the
197 BGTC architecture (Fig. 3a), and with channel length (L) of 10 μm and width (W) of 50 μm (Fig. 3b).
198 The transistors use a molybdenum (Mo) gate electrode deposited onto glass substrate and then
199 patterned through photolithography, a spin-coated hafnium oxide (HfO₂, ~40 nm in thickness) gate
200 dielectric, a ~40 nm perovskite channel spin-coated onto HfO₂ and post-treated with TMA-PF₆ (with
201 x% w/w in CB, where x = 0,1,2 and 4) followed by gold (Au) source (S) and drain (D) contacts using
202 electron beam evaporation through a shadow mask (more details are included in Methods). We

203 measured a stable capacitance of $\sim 386 \text{ nF cm}^{-2}$ in Mo/HfO₂/Au metal-insulator-metal (MIM)
204 structures. The capacitance remained nearly unaffected when the MIM structures were dipped into
205 the DMF:DMSO (4:1) solvent for 120 min (Supplementary Fig. 16), an indication that the gate
206 dielectric is not affected by the spin-coating of the perovskite channel.

207 The transistors were measured in a continuous mode inside a nitrogen filled glove box, in the
208 dark at room temperature (Supplementary Video 1 and 2). Best performance was obtained when
209 treating the perovskite surface with 2% w/w TMA-PF₆ solutions (Fig. 3c, Supplementary Fig. 17).
210 The corresponding performance parameters of these TFTs, in particular, the electron mobilities,
211 current on/off ratios, threshold voltages (V_{TH}) and subthreshold swing (SS) (calculated using eq. (1)
212 and (2) in Methods), are summarized in Supplementary Table 7. Representative T-FAPbI₃ TFTs
213 demonstrate a record high μ_e up to $25.15 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ (Fig. 3c), a $I_{\text{on}}/I_{\text{off}}$ ratio of 10^7 , a V_{TH} of 1.84 V
214 and a SS of 240 mV dec^{-1} . The T-FAPbI₃ transistors present remarkable reproducibility
215 (Supplementary Fig. 18), ultra-low leakage current and zero hysteresis as indicated by the good
216 matching of transfer and output characteristics recorded under forward and reverse scan (FS/RS) (Fig.
217 3d,e). Additionally, they show low operation voltage combined with a transconductance (g_m , which is
218 the main transistor parameter that governs signal amplification) of up to 35 mS mm^{-1} (Fig. 3f,g).
219 Although extremely high transconductance above 500 mS mm^{-1} has been reported for n-channel Si
220 MOSFETs⁴³, the value reported herein is comparable with best performing solution-processed
221 organic electrochemical transistors⁴⁴. The TFT N_{trap} was also calculated for α -FAPbI₃ and T-FAPbI₃
222 channels. Using eq. (3) in Methods, an interface trap density of N_{it} of $8.7 \times 10^{11} \text{ cm}^{-2} \text{ eV}^{-1}$ was
223 calculated for T-FAPbI₃ transistor, which is half of that found for the $1.64 \times 10^{12} \text{ cm}^{-2} \text{ eV}^{-1}$ of α -
224 FAPbI₃ TFT. This is indicative of the enhanced overall quality of the perovskite/HfO₂ interface in
225 the TMA-PF₆ treated transistor as a result of the reduced density of undercoordinated Pb present at
226 the perovskite/HfO₂ interface⁴⁵.

227 Furthermore, we assessed the stability of the optimized perovskite TFTs, which is important
228 for their practical use. To avoid the oxygen-induced aging of our devices, the stability tests were
229 carried out inside nitrogen filled glove box. During the stability study, their electrical properties were
230 continuously measured for 150ks (nearly 42 hours) upon the application of positive bias stress (PBS).
231 Fig. 3h shows the corresponding transfer curves with a positive threshold voltage (V_{TH}) shift whereas
232 Fig. 3i shows the shift of normalized drain current of the transistor after a constant bias stress with a
233 V_{GS} of +5 V for 150ks. It is observed that for the first 90ks (about 25 hours), the transistor maintains
234 nearly 80% of its initial current with only a 0.45 increase in V_{TH} . The transistor is even more stable
235 under negative bias stress (NBS). Fig. 3j shows the evolution of the transfer curve under the NBS
236 with a V_{GS} of -5 V for 150ks. The negligible threshold voltage shift (ΔV_{TH}) and no sign of

237 deterioration of the perovskite TFT under NBS indicate few defects in the bulk and surface of the
238 channel layer and no screening of the applied gate potential. Moreover, the on/off switching
239 characteristics of the optimized TFTs showed a constant current response with clear on and off states
240 for 5000 cycles (Supplementary Fig. 19), satisfying the requirement as a switching unit, which is a
241 critical figure of merit for applications⁴⁶.

242 Such remarkable stability and high electron mobility are the results of our strain relaxation
243 and multidentate anchoring approach that benefits the perovskite microstructure and “cleans” the
244 bulk and surface from undesired structural defects. Notably, a champion electron mobility of 33 cm^2
245 $\text{V}^{-1} \text{ s}^{-1}$ was obtained upon further process optimization (application of a short post-annealing at $50 \text{ }^\circ\text{C}$,
246 2 min after the deposition of the TMA-PF₆, Fig. 3k,l and Supplementary Fig. 20). Our results
247 indicate the crucial role of optimizing the perovskite channel in terms of lattice relaxation and
248 passivation of structural imperfections to the achievement of record performance in perovskite
249 transistors (Supplementary Table 8). We also fabricated n-type transistors using traditional Si/SiO₂
250 wafer gate/gate dielectric substrates. Despite the low dielectric constant of SiO₂ (3.9 compared to 25
251 of HfO₂), we achieved a μ_e of $18 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ with an $I_{\text{on}}/I_{\text{of}}$ current of 10^8 (Supplementary Fig. 21),
252 demonstrating the universality of our approach.

253

254 **Fabrication of all-perovskite logic circuits**

255 We next fabricated circuits with the optimized T-FAPbI₃ n-channel transistors to demonstrate their
256 potential for use in logic electronics. For these circuits, we used Mo bottom gate, solution processed
257 HfO₂ as the dielectric layer, perovskite channels and Au S/D electrodes. All individual TFTs were of
258 the same channel length ($10 \text{ }\mu\text{m}$) and width ($50 \text{ }\mu\text{m}$). An integrated circuit consisted of 326 n-channel
259 perovskite TFTs, 40 unipolar inverters and 10 ring oscillators (RO) (Supplementary Fig. 22). An
260 inverter circuit is a basic logic element that produces an output logic level opposite to its input logic
261 level. Optical images of the unipolar inverter both in low and high magnification, along with the
262 equivalent electronic circuit are presented in Fig. 4a and Fig. 4b, respectively. These unipolar
263 inverter consists of two n-channel perovskite TFTs for both load and drive-TFT devices. Load-TFT
264 operates as a resistor whereas drive-TFT can be controlled from the OFF to ON state so that input
265 and output can be inverted. The gate electrode connects to the drain electrode of the load-TFT, which
266 operates in the saturation region.

267 When the drive-TFT is in the cut-off (for a low input voltage, V_{IN}), no current flows in the
268 circuit and the load-TFT pulls the output voltage (V_{OUT}) toward the supply voltage (V_{DD}). We
269 achieved rectangular-shaped voltage-transfer characteristics of the n-type unipolar inverter measured
270 at room temperature under V_{DD} from 0 to 4V and V_{IN} ranging from -2 to +2 V (Fig. 4c). The

271 magnitude of the V_{OUT} obtained from the inverter is found to be the same as V_{DD} , as expected for
272 low V_{IN} as the drive TFT operates in the cut-off region. From the negative slope ($-\partial V_{OUT}/-\partial V_{IN}$) of
273 the voltage transfer characteristics we calculated the gain value of 16.76 for a V_{DD} of 4 V (Fig. 4d
274 and Supplementary Table 9).

275 Ring oscillators were also fabricated using n-channel perovskite TFTs. Fig. 4e an optical
276 image (along with a zoom in) of the 11-stage ROs fabricated to identify how fast all-perovskite TFTs
277 could work in an integrated circuit. The output of each inverter is connected to the input of the next
278 one except for the 11th stage inverter, which feeds back the input of the first inverter in the RO,
279 resulting in oscillations in the circuit. It is noticed that the oscillation frequency increases as a
280 function of the supply voltage (Fig. 4f). The inset in Fig. 4f presents the output sinusoidal waveform
281 obtained from the RO upon the application of a supply voltage V_{DD} of 4V and oscillation frequency
282 of 78.5 kHz. Furthermore, the corresponding propagation delay per stage was found 581.2 ns at a
283 V_{DD} of 4 V. These results suggest that perovskite TFT is a viable replacement for conventional
284 electronics in logic circuit applications.

285

286 **Conclusions**

287 We have reported high-performance n-channel perovskite TFTs based on the stabilized alpha phase
288 of the FAPbI₃ perovskite semiconductor. Through the application of a combined strain relaxation and
289 multidentate anchoring approach, we modify the perovskite film morphology/crystallinity, while
290 cleaning the bulk and surface from undesired defects, such as undercoordinated Pb. The optimized
291 TFTs using solution-processed HfO₂ gate dielectrics show average field-effect electron mobilities of
292 25.15 cm² V⁻¹ s⁻¹ and maximum field-effect electron mobilities of 33 cm² V⁻¹ s⁻¹, with no hysteresis
293 and high stability under positive and negative bias stresses for 150,000 seconds. Our n-type
294 transistors were used to create all-perovskite circuits in the form of unipolar inverters and 11-stage
295 ring oscillators. The results could lead to the development of high-performance integrated circuits
296 based on low-cost all-solution processes.

297

298 **Methods**

299 **Materials**

300 *N,N*-dimethylformamide (DMF, anhydrous 99.8%, Aldrich), dimethyl sulfoxide (DMSO, anhydrous
301 >99.5%, Aldrich), 2-methoxy ethanol (2ME anhydrous, 99.8%, Aldrich), formamidinium acetate salt
302 (99%, Alfa Aesar), hydroiodic acid (HI, 57 wt% in water, Aldrich), lead iodide (PbI₂ 99.99% PbI₂,
303 TCI), methylammonium chloride (MACl, 98%, Acros Organics), tetramethylammonium
304 hexafluorophosphate (TMA-PF₆, 98%, Aladdin), diethyl ether (DE extra pure grade, Duksan),

305 tetrahydrofuran (THF, anhydrous 99.9% Aldrich), chlorobenzene (CB, anhydrous, 99.8%, Aldrich),
306 Au (99.999%, Kurt J. Lesker). All chemicals were used without any further purification process.

307 **Preparation of precursor solutions**

308 For the fabrication of the perovskite layer, the process was performed in at controlled environment
309 (temperature of 20 °C inside a nitrogen-filled glove box). FAI was prepared by mixing 25 g
310 formamidine acetate with 50 ml hydroiodic acid in a 500 ml round-bottomed flask with vigorous
311 stirring. A yellowish powder was obtained by evaporating the solvent at 80 °C for 1 h in a vacuum
312 evaporator. The powder was dissolved in ethanol and precipitated using diethyl ether. This step was
313 repeated three times until white powder was obtained, which was recrystallized from ethanol and
314 diethyl ether in a refrigerator. After recrystallization, the resulting powders were collected and dried
315 at 60 °C for 24 h. FAPbI₃ precursor solution was prepared by mixing 368.8 mg PbI₂ with 137.58 mg
316 FAI in 1mL of DMF:DMSO (4:1). The solution was stirred for 1 h to 70 °C. For FAPbI₃:MACl_x%,
317 MACl was added in the range 0-55 mol% (i.e., for FAPbI₃:MACl₃₅%, 37 mg of MACl were added
318 in 1 mL of FAPbI₃) and the derived solution was stirred for 10 min without heating. For THF
319 dilution, the solution was mixed with x % per volume THF i.e., add 300 μL THF into 100 μL
320 perovskite solution for 1:3%v/v dilution, followed by stirring without heating.

321

322 **Device fabrication**

323 Typical bottom gate top contact devices were fabricated on glass substrates using HfO₂ as the gate
324 dielectric. The substrates were first cleaned sequentially with acetone, ethanol and isopropyl alcohol
325 (IPA) in an ultrasonic system for 15 min. First, a 40 nm Mo layer was sputtered on the substrate and
326 lithographically patterned to form the gate electrode. The gate dielectric was deposited from a 0.2 M
327 HfO₂ precursor solution prepared by dissolving hafnium chloride (HfCl₄) in 2 methoxy ethanol. The
328 precursor solution was stirred in an N₂ atmosphere for two hours and filtered using a 0.45 μm
329 poly(tetrafluoroethylene) (PTFE) syringe filter before being used. HfO₂ was deposited by spin-
330 coating 50 μl of the solution at 3500 rpm for 30 s. After spin-coating, the film was cured at 200 °C
331 on a hot plate for 5 min and then ozone treated for 10 min using a high-pressure mercury UV lamp
332 while the sample was kept at 100 °C. Finally, the thin films were annealed at 300°C for two more
333 hours in an air furnace. Prior perovskite deposition, contact holes were made by dry etching and
334 patterned it to form “banks” for the perovskite semiconductor. For the perovskite channel preparation,
335 70 μL of the THF diluted (1:3 %v/v) FAPbI₃:MACl₃₅% precursor solution was spread over the
336 HfO₂ layer at 6000 rpm for 50 s with 0.1 s ramping. Prior to that, the HfO₂ substrate was treated with
337 UV ozone for 30 min. During the spin-coating, antisolvent treatment was applied by dripping 1mL
338 DE after spinning for 10 s. The perovskite film was then dried on a hot plate at 150 °C for 10 min.

339 For surface treatment, 50 μL of TMA-PF₆ (2 % w/w in CB) was spin-coated onto the perovskite film
 340 at 5000 rpm for 30 s with no thermal annealing. The S/D Au electrodes were next deposited through
 341 a shadow mask under a high vacuum (10^{-7} torr) using a thermal evaporator placed in a N₂-filled
 342 glove box. The channel width and length of all TFTs were fixed at 10 and 50 μm , respectively. For
 343 the n-inverter and ring oscillator, Mo deposited and patterned on glass were used as the bottom gate,
 344 solution processed HfO₂ as the dielectric layer, THF diluted T-FAPbI₃ (FAPbI₃:MACl35% surface
 345 treated with TMA-PF₆) as the n-channel and evaporated gold through a shadow mask as electrodes.

346

347 **Transistor measurements**

348 The transistor, inverter and ring oscillator measurements were performed at room temperature in the
 349 dark inside a nitrogen-filled glove box. The perovskite TFTs were measured using an Agilent 4156C
 350 semiconductor parameter analyzer in the continuous bias mode. The transfer curves were measured
 351 at a drain voltage (V_{DS}) of 0.1 V by sweeping the gate voltage (V_{GS}) from -5 to $+5$ V. The output
 352 curve was measured at a V_{GS} of 5 V with a step of 1 V by sweeping the V_{DS} from 0 to $+5$ V. The
 353 field-effect mobility in the saturation region ($V_{DS} \geq V_{GS} - V_{TH}$) was obtained by using the equation,

$$354 \quad I_{DS} = \frac{1}{2} \frac{W}{L} \mu_{sat} C_{ox} (V_{GS} - V_{TH})^2 \quad (1)$$

355 where the threshold voltage (V_{TH}) and saturation mobility were determined from the x-axis intercept
 356 and linear part of the $(I_{DS})^{1/2}$ vs V_{GS} plot by the linear extrapolation method.

357 The subthreshold swing (SS) was obtained from the linear region of the log (I_{DS}) versus V_{GS} fit by
 358 using the following equation:

$$359 \quad SS = \frac{dV_{GS}}{d(\log I_{DS})} \quad (2)$$

360 Here, interfacial trap density (N_{it}) was obtained from the equation:

$$361 \quad N_{it} = \left| \frac{(SS \log(e))}{\frac{KT}{q}} \right| \frac{C_{ox}}{q} \quad (3)$$

362 where I_{DS} , W/L , μ_{sat} , C_{ox} , V_{GS} , and V_{TH} are the drain current, channel width/length, saturation
 363 mobility, capacitance of the gate oxide, gate voltage, and threshold voltage, respectively.

364

365 **Computational details**

366 To simulate the effect of TMA-PF₆ incorporation into the bulk of FAPbI₃-MACl35% a structure
 367 containing ca 2% of TMA-PF₆ was constructed, using a 4x4x4 supercell of the initial perovskite
 368 structure. The adsorption of TMA-PF₆ on the perovskite surface was modeled on the (001) surface
 369 slab. The structures were visualized using VESTA⁴⁷ and VMD⁴⁸ programs. The effective masses of
 370 holes and electrons were computed for the 2x2x2 supercell structures containing either 0% or 12.5%

371 TMA-PF₆, which were both relaxed in terms of cell parameters and atomic positions using PBE
372 density functional employing 40 Ry and 280 Ry kinetic energy and charge density cutoffs and fully
373 relativistic ultrasoft pseudopotentials⁴⁹, as implemented in Quantum Espresso code⁵⁰. More details
374 are included in Supplementary Information.

375

376 **Nuclear magnetic resonance (NMR)**

377 Proton nuclear magnetic resonance (¹H-NMR) and fluorine nuclear magnetic resonance (¹⁹F-NMR)
378 spectra were obtained with a Bruker AVANCE 400 MHz instrument in deuterated DMSO. Nine
379 series of titration experiments were carried out, by varying the concentration of a certain substance in
380 the presence of one or a mixture of “initial” substance(s), whose concentration remained stable. The
381 amount of the initial substance(s) was set to 0.048 mmol, whereas the amount of the substance
382 gradually added was set at: 0 equivalents, 0.35 equivalents, 0.55 equivalents, 0.75 equivalents, 1.0
383 equivalents, and 2.0 equivalents, respectively (equivalents are defined as molar equivalents with
384 respect to the initial substance(s)). Thus, for each series of measurements, six ¹H (and ¹⁹F where
385 relevant) NMR spectra were obtained. The titration series that were studied are: (a) addition of x
386 equivalents of MACl to 0.048 mmol of FAI, (b) addition of x equivalents of MACl to 0.048 mmol of
387 PbI₂, (c) addition of x equivalents of FAI to 0.048 mmol of TMA-PF₆, (d) addition of x equivalents
388 of MACl to 0.048 mmol of TMA-PF₆, (e) addition of x equivalents of PbI₂ to 0.048 mmol of TMA-
389 PF₆, (f) addition of x equivalents of PbI₂ to an equimolar mixture (0.048 mmol each) of FAI and
390 MACl, (g) addition of x equivalents of FAI to an equimolar mixture (0.048 mmol each) of MACl
391 and PbI₂, (h) addition of x equivalents of THF to 0.048 mmol of FAI, and (i) addition of x
392 equivalents of THF to 0.048 mmol of PbI₂. Representative experimental procedure: In a 4 mL vial,
393 0.048 mmol (8.2 mg) of FAI were added, followed by the addition of x mmol (0, 0.35, 0.55, 0.75, 1.0,
394 or 2.0 equivalents) of MACl. To this vial, 0.6 ml of DMSO-d₆ were added, the mixture was
395 mechanically stirred until complete dilution of all solids. The mixture was then analyzed via ¹H- or
396 ¹⁹F-NMR.

397

398 **Other characterisation techniques**

399 The surface of the perovskite films was imaged using a Zeiss LEO 1550 FE-SEM apparatus with a 2
400 kV acceleration voltage. It was ensured that the voltage levels for SEM measurement did not
401 significantly impact the integrity of the perovskite thin films. Atomic force microscopy (AFM)
402 images were captured using an XE7 microscope (Park Systems) by operating in tapping mode. The
403 crystallinity of perovskite films was investigated using an X-ray Siemens D-500 606 diffractometer.
404 XPS measurements were performed by a monochromatic Al K α X-ray source (h ν = 1486.6 eV) using

405 a SPECS PHOIBOS 150 electron energy analyser with a total energy resolution of 500 meV.
406 Conductive silver paint was used to connect the sample surface to the holder to avoid charge
407 accumulation during XPS measurements. All the films were deposited on Si substrates in the same
408 way as used for FET devices. UPS measurements were carried out using an ultralow-background,
409 high-sensitivity UPS apparatus with a hemispherical electron energy analyzer (MBS A-1),
410 monochromatic He I α ($h\nu = 21.22$ eV) and Xe I ($h\nu = 8.44$ eV) radiation sources. The UPS spectra
411 were recorded at normal emission with an acceptance angle of $\pm 18^\circ$ in transmission mode. A bias of
412 -5 V was applied to the substrate to obtain the secondary electron cut-off. The spectral energy
413 resolution was estimated to be less than 30 meV measured from the gold Fermi edge. PL
414 measurements were performed using a confocal microscope (WITec, Alpha RAS system). A fiber-
415 coupled 405-nm continuous-wave laser (Coherent, CUBE) was focused onto the sample using 40 \times
416 objective. The average power of the laser at its focal point was 0.5 μ W. PL of the sample was
417 collected in the reflection geometry from the same objective, while the excitation laser beam from
418 the reflection was blocked using a 415-nm long-pass filter. Time-resolved photoluminescence (TRPL)
419 spectra were measured with an FS5 spectrofluorometer from Edinburgh Instruments. A 478.4 nm
420 laser was used as an excitation source. UV–Visible absorption spectra were recorded using a Scinco
421 S-4100 spectrometer. Fourier-transform infrared (FTIR) spectroscopy was performed using a Bruker
422 Tensor 27 spectrophotometer with a DTGS detector. Two dimensional grazing incidence X-ray
423 diffraction (2D-GIXRD) results were obtained by using Xenocs Xeuss 2.0 instrument with a Cu X-
424 ray source, and the wavelength and the incident angle of the X-ray beam are 0.154 nm and 0.2° ,
425 respectively. Depth analysis X-ray photoelectron spectroscopy (XPS) was conducted with an Axis
426 Ultra, Kratos-Analytical Ltd apparatus.

427

428 **Data availability**

429 Source data are provided with this paper. Additional data related to this work are available from the
430 corresponding authors upon request.

431

432 **Code availability statement**

433 All codes (software) used in the calculation and visualization are publicly available and the condition
434 of their usage in the publication is an appropriate citation.

435

436 **References**

- 437 1. Kagan, C., Mitzi, D. & Dimitrakopoulos, C. Organic-inorganic hybrid materials as
438 semiconducting channels in thin-film field-effect transistors. *Science* **286**, 945–947 (1999).

- 439 2. Lin, Y.-H., Pattanasattayavong, P. & Anthopoulos, T. D. Metal-halide perovskite transistors for
440 printed electronics: challenges and opportunities. *Adv. Mater.* **29**, 1702838 (2017).
- 441 3. Paulus, F., Tyznik, C., Jurchescu, O. D. & Vaynzof, Y. Switched-on: progress, challenges, and
442 opportunities in metal halide perovskite transistors. *Adv. Funct. Mater.* **31**, 2101029 (2021).
- 443 4. Zhu, H., Liu, A. & Noh, Y.-Y. Recent progress on metal halide perovskite field-effect transistors.
444 *J. Inf. Disp.* **22**, 1–12 (2021).
- 445 5. She, X. J. et al. A solvent-based surface cleaning and passivation technique for suppressing ionic
446 defects in high-mobility perovskite field-effect transistors. *Nat. Electron.* **3**, 694–703 (2020).
- 447 6. Matsushima, T. et al. Solution-processed organic–inorganic perovskite field-effect transistors
448 with high hole mobilities. *Adv. Mater.* **28**, 10275–10281 (2016).
- 449 7. Gao, Y. et al. Highly stable lead-free perovskite field-effect transistors incorporating linear π -
450 conjugated organic ligands. *J. Am. Chem. Soc.* **141**, 15577–15585 (2019).
- 451 8. Zhu, H. et al. Perovskite and conjugated polymer wrapped semiconducting carbon nanotube
452 hybrid films for high-performance transistors and phototransistors. *ACS Nano* **13**, 3971–3981
453 (2019).
- 454 9. Zhu, H. et al. High-performance and reliable lead-free layered-perovskite transistors. *Adv. Mater.*
455 **32**, 2002717 (2020).
- 456 10. Liang, A. et al. Ligand-driven grain engineering of high mobility two-dimensional perovskite
457 thin-film transistors. *J. Am. Chem. Soc.* **143**, 15215–15223 (2021).
- 458 11. Shao, S. et al. Field-effect transistors based on formamidinium tin triiodide perovskite. *Adv.*
459 *Funct. Mater.* **31**, 2008478 (2021).
- 460 12. Yusoff, A. R. B. M. et al. Ambipolar triple cation perovskite field effect transistors and inverters.
461 *Adv. Mater.* **29**, 1602940 (2017).
- 462 13. Kim, H. P. et al. A hysteresis-free perovskite transistor with exceptional stability through molecular
463 cross-linking and amine-based surface passivation. *Nanoscale*, **12**, 7641–7650 (2020).
- 464 14. Zhu, H. et al. High-performance hysteresis-free perovskite transistors through anion engineering.
465 *Nat. Commun.* **13**, 1741 (2022).
- 466 15. Li, F. et al. Ambipolar solution-processed hybrid perovskite phototransistors. *Nat. Commun.* **6**,
467 8238 (2015).
- 468 16. Liu, A. et al. High-performance inorganic metal halide perovskite transistors. *Nat Electron* **5**, 78–
469 83 (2022).
- 470 17. Zhu, H. et al. Tin perovskite transistors and complementary circuits based on A-site cation
471 engineering. *Nat. Electron.* **6**, 650–657 (2023).

- 472 18. Yu, W. et al. Single crystal hybrid perovskite field-effect transistors. *Nat. Commun.* **9**, 5354
473 (2018).
- 474 19. Liu, A. et al. Modulation of vacancy-ordered double perovskite Cs₂SnI₆ for air-stable thin-film
475 transistors. *Cell Reports Physical Science* **3**, 100812 (2022).
- 476 20. Senanayak, S.P. et al. Charge transport in mixed metal halide perovskite semiconductors. *Nat.*
477 *Mater.* **22**, 216–224 (2023).
- 478 21. Wang, G. et al. Wafer-scale growth of large arrays of perovskite microplate crystals for
479 functional electronics and optoelectronics. *Sci. Adv.* **1**, e1500613 (2015).
- 480 22. Li, D. et al. Size-dependent phase transition in methylammonium lead iodide perovskite
481 microplate crystals. *Nat. Commun.* **7**, 11330 (2016).
- 482 23. Senanayak, S. P. et al. Understanding charge transport in lead iodide perovskite thin-film field-
483 effect transistors. *Sci. Adv.* **3**, e1601935 (2017).
- 484 24. Jeong, J. et al. Pseudo-halide anion engineering for α -FAPbI₃ perovskite solar cells. *Nature* **592**,
485 381–385 (2021).
- 486 25. Min, H. et al. Efficient, stable solar cells by using inherent bandgap of α -phase formamidinium
487 lead iodide. *Science* **366**, 749–753 (2019).
- 488 26. Chin, X. Y., Cortecchia, D., Yin, J., Bruno, A. & Soci, C. Lead iodide perovskite light-emitting
489 field-effect transistor. *Nat. Commun.* **6**, 7383 (2015).
- 490 27. Kim, M. et al. Methylammonium chloride induces intermediate phase stabilization for efficient
491 perovskite solar cells. *Joule* **3**, 2179–2192 (2019).
- 492 28. Cheng, Z. et al. How to report and benchmark emerging field-effect transistors. *Nat. Electron.* **5**,
493 416–423 (2022).
- 494 29. Suzuki, A., Oku, T. Effects of mixed-valence states of Eu-doped FAPbI₃ perovskite crystals studied by
495 first-principles calculation. *Mater. Adv.* **2**, 2609–2616 (2021).
- 496 30. Li, Z. et al. Ammonia for post-healing of formamidinium-based Perovskite films. *Nat. Commun.*
497 **13**, 4417 (2022).
- 498 31. Kieslich, G., Sun, S. & Cheetham, A. K. Solid-state principles applied to organic–inorganic
499 perovskites: new tricks for an old dog. *Chem. Sci.* **5**, 4712–4715 (2014).
- 500 32. Kim, M. et al. Methylammonium chloride induces intermediate phase stabilization for efficient
501 perovskite solar cells. *Joule* **3**, 2179–2192 (2019).
- 502 33. Gerhard, M. et al. Microscopic insight into non-radiative decay in perovskite semiconductors
503 from temperature-dependent luminescence blinking. *Nat. Commun.* **10**, 1698 (2019).

- 504 34. Van Gompel, W.T.M. *et al.* Degradation of the formamidinium cation and the quantification of
505 the formamidinium–methylammonium ratio in lead iodide hybrid perovskites by nuclear
506 magnetic resonance spectroscopy. *J. Phys. Chem. C* **122**, 8, 4117–4124 (2018).
- 507 35. Kubicki, D.J. *et al.* NMR spectroscopy probes microstructure, dynamics and doping of metal
508 halide perovskites. *Nat. Rev. Chem.* **5**, 624–645 (2021).
- 509 36. Wu, X. Trap states in lead iodide perovskites. *J. Am. Chem. Soc.* **137**, 5, 2089–2096 (2015).
- 510 37. Wu, W.-Q. *et al.* Reducing surface halide deficiency for efficient and stable iodide-based
511 perovskite solar cells. *J. Am. Chem. Soc.* **142**, 8, 3989–3996 (2020).
- 512 38. Kim, H.S., Park, N.G. Importance of tailoring lattice strain in halide perovskite crystals. *NPG Asia*
513 *Mater.* **12**, 78 (2020).
- 514 39. Qiao, H. W. *et al.* A Gradient heterostructure based on tolerance factor in high-performance
515 perovskite solar cells with 0.84 fill factor. *Adv. Mater.* **31**, 1804217 (2019).
- 516 40. Hu, J. *et al.* Tracking the evolution of materials and interfaces in perovskite solar cells under an
517 electric field. *Commun. Mater.* **3**, 39 (2022).
- 518 41. Cacovich, S. *et al.* Imaging and quantifying non-radiative losses at 23% efficient inverted
519 perovskite solar cells interfaces. *Nat. Commun.* **13**, 2868 (2022).
- 520 42. Lin, Y.-H. *et al.* A piperidinium salt stabilizes efficient metal-halide perovskite solar cells.
521 *Science* 369, 96–102 (2020).
- 522 43. Horiguchi, S., Kobayashi, T., Miyake, M., Oda, M. & Kiuchi, K Extremely high
523 transconductance (above 500 mS/mm) MOSFET with 2.5 nm gate oxide. *1985 International*
524 *Electron Devices Meeting, IEDM* 10.1109/IEDM.1985.191088 (1985).
- 525 44. Khodagholy, D. *et al.* High transconductance organic electrochemical transistors. *Nat Commun* **4**,
526 2133 (2013).
- 527 45. Lin, Y.H. *et al.* Hybrid organic–metal oxide multilayer channel transistors with high operational
528 stability. *Nat Electron.* **2**, 587–595 (2019).
- 529 46. Kovalenko, M. V., Protesescu, L. & Bodnarchuk, M. I. Properties and potential optoelectronic
530 applications of lead halide perovskite nanocrystals. *Science* **358**, 745–750 (2017).
- 531 47. Momma, K.; Izumi, F., VESTA 3 for three-dimensional visualization of crystal, volumetric and
532 morphology data. *Journal of Applied Crystallography* **44**, 1272–1276 (2011).
- 533 48. Humphrey, W.; Dalke, A.; Schulten, K., VMD: Visual molecular dynamics. *Journal of*
534 *Molecular Graphics* **14** (1), 33–38 (1996).
- 535 49. Rappe, A. M.; Rabe, K.M.; Kaxiras. E. & Joannopoulos, J. D. Optimised pseudopotentials. *Phys.*
536 *Rev. B* **41**, 1227 (R) (1990).

537 50. Giannozzi, P. et al. QUANTUM ESPRESSO: a modular and open-source software project for
538 quantum simulations of materials. *J. Phys. Condens. Matter.* **21**(39):395502 (2009).

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541

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550

551 **Contributions**

552 A.R.B.M.Y., M.V., R.N.B. and M.K.N. conceived and designed the project. M.V., M.K.N. and
553 A.R.B.M.Y. supervised the project and led the collaboration efforts. R.N.B., A.S., A.V. and M.T.
554 characterized the materials. V.P. performed the XRD studies. L.P.Z. and G.C.V. performed the NMR
555 measurements and analysis. K.A. conducted the numerical simulations in perovskite diodes. R.N.B.
556 and T.A. completed the electrical device fabrication, performed the luminescence measurements and
557 conducted the experimental validation. J.W. and X.B. performed depth XPS and GIWAXS studies.
558 O.A.S., M.A.S. and H.U. performed the theoretical calculations. M.V. wrote the manuscript. M.K.N.
559 and A.R.B.M.Y. edited the manuscript. All the authors participated in the discussion and analysis of
560 the manuscript.

561

562 **Competing interests**

563 The authors declare no competing interests.

564

565 **Inclusion & Ethics statement**

566 The authors have followed the recommendations set out in the Global Code of Conduct for Research
567 in Resource-Poor Settings when designing, executing and reporting their research.

568

569 **Fig. 1 Managing surface morphology, crystallinity and crystal orientation.** **a**, SEM top view
570 images of FAPbI₃ perovskite films **a**, pristine, **b**, added 35% per molar MACl (denoted as α -FAPbI₃),

571 **c**, diluted with THF (1:3 % v/v) and **d**, surface treated with TMA-PF₆ (denoted as T-FAPbI₃). **e**,
572 XRD peaks of the same FAPbI₃, α -FAPbI₃ before and after THF dilution and TMA-PF₆ treated α -
573 FAPbI₃ films. The position of (100) plane peak at different perovskite films along with the PbI₂ and
574 δ -FAPbI₃ perovskite phase related peaks, present only in the pattern of pristine perovskite film, are
575 shown. GIWAXS patterns of **f**, pristine FAPbI₃, α -FAPbI₃ (added 35% MACl), **g**, before THF
576 dilution, and **h**, after THF dilution, and of **i**, T-FAPbI₃.

577

578 **Fig. 2 Strain relaxation and multidentate anchoring.** **a**, Changes in the ¹H-NMR spectra of FAI
579 (0.048 mmol) by gradual addition of MACl (0-2 equiv). **b**, Changes in the ¹H-NMR spectra of PbI₂
580 (0.048 mmol) by gradual addition of MACl (0-2 equiv). **c**, Changes in the ¹⁹F-NMR spectra of TMA-
581 PF₆ (0.048 mmol) by gradual addition of FAI (0-2 equiv). **d**, Changes in the ¹⁹F-NMR spectra of
582 TMA-PF₆ (0.048 mmol) by gradual addition of PbI₂ (0-2 equiv). **e**, Pb 4f and **f**, I 3d XPS core level
583 peaks of pristine FAPbI₃, α -FAPbI₃ (including 35% MACl and THF diluted) and T-FAPbI₃, which is
584 the TMA-PF₆ (2% w/w in CB) surface treated sample. **g**, Schematic illustration of the proposed
585 strain relaxation by incorporating MA cations within the perovskite crystal for compensating the
586 lattice strain. Also, illustration of the multidentate anchoring of TMA-PF₆ at the grain boundaries
587 and/or surfaces of FAPbI₃ perovskite. **h**, DFT calculations on I-F and hydrogen bonding modes of
588 the TMA-PF₆ in the bulk of perovskite. **i**, DFT results on halogen and hydrogen bonding modes of
589 the TMA-PF₆ on the perovskite surface.

590

591 **Fig. 3 The electrical performance and stability of n-channel T-FAPbI₃ perovskite TFTs.** **a**,
592 Schematic diagram of the bottom gate top contact (BGTC) perovskite TFT. **b**, An optical image of
593 the perovskite transistor with a channel length (L) of 10 μ m and width (W) of 50 μ m. **c**, The field
594 effect electron mobility (μ_e) of TFTs based on channels fabricated from FAPbI₃:MACl35%
595 precursors with different x (x = 0, 1, 2 and 4) % w/w concentrations of TMA-PF₆ for surface
596 treatment. **d**, The transfer characteristics and **e**, the output characteristics of representative n-channel
597 α -FAPbI₃ perovskite transistors for different gate bias in forward (FS) and reverse scan (RS). **f**, The
598 electron mobility (μ_e) as a function of gate voltage of the TFT. The transfer curve was measured by
599 sweeping V_{GS} from -5 to +5 V at V_{DS} of 0.1 V. **g**, Transconductance of the perovskite TFT. **h**,
600 Continuous measurements of transfer curves in FS and RS of a representative T-FAPbI₃ TFT under
601 positive-bias stress (V_{GS} = +5 V, V_{DS} = +0.1 V) for different durations up to 150ks. **i**, The shift of
602 normalized drain current of the transistor after a constant positive-bias stress for 150ks. **j**, Transfer
603 curves under the negative-bias stress (V_{GS} = -5 V, V_{DS} = -0.1 V) for durations up to 150ks. **k**,
604 Transfer and **l**, output characteristics of a champion perovskite TFT.

605

606

607 **Fig. 4 All-perovskite unipolar inverter and 11-stage ring oscillator. a**, An optical image (left) and
608 zoom-in (right) of the unipolar inverter. **b**, The equivalent electronic circuit. **c**, Voltage transfer
609 characteristics (VTC) and **d**, voltage gain ($-\partial V_{\text{OUT}}/\partial V_{\text{IN}}$) of α -FAPbI₃ perovskite TFT inverter with
610 a different V_{DD} from 1 to 4 V. **e**, An optical microscopy image of the integrated ring oscillators (left)
611 and zoom-in (right) to an individual ring oscillator. **f**, Oscillation frequency as a function of supply
612 voltage (V_{DD}) of the α -FAPbI₃ ring oscillator. The inset shows the output voltage of the T-FAPbI₃
613 ring oscillator at a frequency of 78.5 kHz.

614







