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Homogeneous bidirectional locking enables reinforced buried interface for high-resolution perovskite X-ray imagers

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Abstract

The large-scale heterogeneous integration of optoelectronic devices is frequently constrained by mismatches in electronic and mechanical properties, as well as the accumulated stress across the integrated interface. This challenge is particularly acute for solution-processed polycrystalline halide perovskite thick films, which, despite their considerable potential for X-ray flat-panel detectors, are hindered by a defective and fragile buried interface that compromises device integrity and leads to poor and non-uniform charge collection. We establish a dense, seamless, and strongly adherent buried interface with markedly enhanced charge extraction by intercalating a vertically-oriented, robust, and homogeneous bidirectional locking interlayer between every single thin-film-transistor pixel and the polycrystalline perovskite absorbers. This strategy improves tensile strength by 61% and substantially augments tolerance to thermal shock cycling between 0 and 100 °C, while concurrently enabling a high charge collection efficiency of 96.2 % for the integrated devices under a low electric field. The resulting imager exhibits a substantially reduced photo-response non-uniformity from 28.57 to 4.31% and achieves an imaging spatial resolution of 6.1 lp mm⁻¹, facilitating three-dimensional X-ray computed tomography reconstruction. This work offers a facile and reliable interface-engineering strategy toward highly integrated and high-resolution perovskite X-ray flat-panel imagers, with promising implications for practical low-dose radiography applications.

The growing demands for precision medical diagnostics, alongside artificial intelligence-driven advanced co-packaged optics and semiconductor packaging, have steadily increased the need of X-ray based non-destructive imaging that affords high spatial resolution while minimizing radiation-induced damage ¹⁻⁴. Direct-type X-ray detectors are particularly advantageous in this regard, as their simple signal conversion pathway and electric-field-driven carrier transport enable superior sensitivity and imaging resolution ⁵. In recent years, semiconductive metal halide perovskites have emerged as transformative candidates for next-generation X-ray imagers owing to their high X-ray absorption coefficient, long carrier diffusion length, high mobility-lifetime ($\mu\tau$) product, and solution processability ⁶⁻¹³. These attributes allow perovskites to effectively overcome the inherent limitations of conventional detection materials such as amorphous selenium (α -Se) and cadmium zinc telluride (CZT).

X-ray imagers typically integrate perovskite films with thin-film transistor (TFT) array panels. However, the substantial mismatch in thermal expansion coefficients between these components poses a significant fabrication challenge ¹⁴. Conventional ultrasonic spraying coating, constrained by cumulative stress buildup during prolonged annealing, can only produce films thinner than 100 micrometers—far below the thickness required for adequate X-ray attenuation ¹⁵. Blade coating, by contrast, has become the mainstream technique due to its excellent process compatibility and scalability ¹⁶⁻¹⁸. Nevertheless, this method introduces particulate features within the films, rapid and non-uniform solvent evaporation during annealing,

and post-coating artifacts, all of which lead to a porous, disordered, and poorly-adhered buried interface (Fig. 1a), resulting in severely compromised charge collection efficiency, pronounced inter-pixel crosstalk, extensive dead pixels, unsatisfactory response uniformity, weak mechanical integrity, and ultimately deteriorated spatial resolution (Fig. 1b) ^{19,20}. Collectively, these critical shortcomings present formidable obstacles to the practical commercialization of perovskite X-ray flat-panel detectors.

Although the perovskite solar cell community has made substantial strides in buried interface optimization to enhance both power conversion efficiency and operational stability, the strategies developed therein are not directly transferable to perovskite X-ray detectors ²¹⁻²³. Tang et al. adopted anisotropic conductive adhesives to improve interfacial bonding between perovskites and substrates, achieving notable gains in mechanical strength ²⁴. Park and Xu et al. developed perovskite-polymer composite buried layers that simultaneously enhance device robustness and carrier transport ^{25,26}. Additional approaches including solvent/gas treatment ^{27,28}, hot-pressing ²⁹, and bottom-seeded crystal growth ³⁰ have also been explored. Despite these advances, the simultaneous achievement of reinforced mechanical properties, oriented carrier transport, and effective dead-pixel elimination remains an outstanding challenge.

Here, we introduce a bidirectional locking strategy that leverages an engineered perovskite interfacial layer featuring vertically oriented, dense columnar microstructures with optimized interfacial stress. This layer establishes a tight and

intimate electrical contact with the TFT pixel electrodes, effectively eliminating bottom-contact dead zones while rectifying the pore defects and poor adhesion inherent to pure blade-coated thick films (Fig. 1c). Moreover, the continuous vertical grain channels within this interlayer afford unobstructed longitudinal carrier transport pathways and promote a quasi-epitaxial coating behavior of the overlying perovskite absorber. As a result, our approach delivers fewer dead pixels, superior signal response uniformity, and remarkably enhanced spatial resolution (Fig. 1d). This work presents a facile, scalable, and universal interfacial and microstructure engineering strategy for high-performance thick perovskite X-ray absorbers, offering a viable pathway toward the industrialization of high-resolution, low-dose perovskite flat-panel X-ray imaging systems.

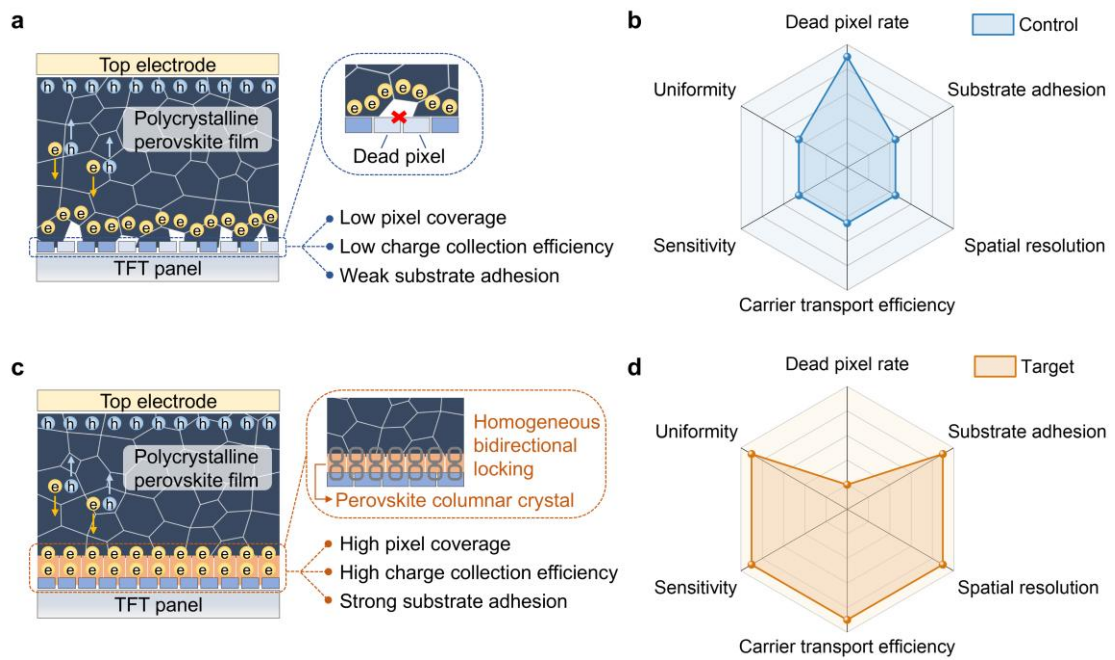


Fig. 1 | Demonstration of bidirectional locking strategy for buried interface optimization and performance improvement for X-ray imaging devices. a, Schematic illustration of the inherent problems in traditional polycrystalline perovskite X-ray FPDs. **b,** Comprehensive performance analysis of traditional polycrystalline

perovskite X-ray FPDs. **c**, Schematic diagram of the comprehensive performance enhancement mechanism of perovskite X-ray FPDs enabled by the vertically oriented bidirectional-locking interface layer. **d**, Comprehensive performance analysis of the optimized perovskite X-ray FPDs with engineered interface layer.

Results

Although blade coating typically produces a defective buried interface, its rapid processing induces relatively low accumulated stress. On the contrary, spray coating with a slow deposition rate introduces substantially higher stress, while its *in-situ* growth mechanism enables the formation of a dense bottom interface³¹. Seeking to capitalize on the complementary advantages of both techniques, we develop a hybrid deposition strategy that effectively circumvents the trade-off between high-quality interfacial contact and sufficient X-ray absorption thickness. Specifically, a thin spray-coated perovskite layer is first deposited to achieve tight interfacial bonding with both the underlying substrate and the subsequently overcoated perovskite film, thereby realizing a robust and homogeneous bidirectional interfacial locking effect. This locked interface then serves as a foundation for subsequent blade-coating deposition of a ~300 μm -thick homogeneous perovskite absorber, a thickness which fully satisfies the X-ray attenuation requirement.

Theoretically, vertically aligned grain structures are advantageous for both enhancing charge transport and improving mechanical robustness, we then tried spray coating high quality columnar perovskite films via solvent engineering. In spray coating, beyond the physical spraying parameters, the precursor solvent coordination

ability and evaporation kinetics play a decisive role in governing nucleation and grain growth behavior, which are essential for achieving a high-quality locked interfacial structure. Common solvents utilized for spray-deposited perovskites can be categorized based on their coordination strength and volatility (Fig. 2a), including non-volatile strong-coordinating dimethyl sulfoxide (DMSO) and dimethylformamide (DMF), non-volatile weak-coordinating γ -valerolactone (GVL), and volatile weak-coordinating 2-methoxyethanol (2-ME). Traditional single-solvent systems based on DMF or DMSO suffer from inefficient solvent removal, requiring elevated substrate temperatures during coating ^{32,33}. Such high-temperature processing inevitably introduces uncontrollable thermal stress and defects, meanwhile readily promotes the formation of undesirable needle-like PbI_2 intermediate phases (Supplementary Fig.1). In comparison, the weakly coordinating GVL system enables rapid crystallization and swift conversion to the perovskite phase, facilitating the fabrication of dense perovskite films ³⁴. However, the unregulated and high-speed nucleation usually yields small, randomly oriented grains without controllable vertical crystallographic texture, which severely limits the formation of ordered columnar perovskite crystals.

To address the limitations of single-solvent formulations, four binary solvent systems with complementary properties were strategically developed for spray-coating MAPbI_3 columnar crystals, namely DMF:2-ME, DMSO:2-ME, GVL:2-ME, and GVL:DMSO. In each binary recipe, the secondary solvent serves as a functional regulator with a fixed volume ratio of 20% (see details in the Methods

section). UV–Vis absorption spectroscopy was performed to characterize the lead-iodide complex speciation in different MAPbI₃ precursor solutions (Fig. 2b). It is well established that Pb²⁺ reacts with I⁻ in polar precursor solutions to form high-order iodoplumbate complexes (e.g., [PbI₃]⁻), which exhibit characteristic absorption signatures in the 350-400 nm range. Strong-coordinating solvents can compete with I⁻ for Pb²⁺ coordination sites, forming low-coordination lead-solvent complexes that manifest as blue-shifted absorption bands ³⁵. As evidenced by the spectral results, both DMF:2-ME and DMSO:2-ME systems show distinct blue-shifted absorption features, which originate from the formation of low-coordinate lead iodide-solvent complexes driven by the strong coordination affinity of DMF and DMSO. In contrast, the GVL:2-ME system, consisting of two weak-coordinating solvents, presents a prominent absorption peak at approximately 370 nm, which is assigned to highly reactive [PbI₃]⁻ species. Such high-order iodoplumbate complexes possess high crystallization activity and tend to rapidly transform into stable perovskite phases during film formation. Notably, the GVL:DMSO system also shows a characteristic [PbI₃]⁻ absorption peak at 370 nm, albeit with a significantly reduced absorbance intensity compared with that of GVL:2-ME. This observation confirms that the introduced DMSO partially occupies the coordination sites of Pb²⁺, which effectively lowers the concentration of highly reactive iodoplumbate species and moderates the overall precursor reactivity.

We subsequently carried out spray-coating of the four precursor solutions under identical substrate temperature conditions (120°C), and the film formation process is

shown in Supplementary Fig.2. Consistent with the solution spectroscopic characteristics, the DMF:2-ME system exhibits sluggish solvent evaporation kinetics, which severely delays the nucleation and crystallization of deposited precursor droplets. This phenomenon is further exacerbated in the DMSO:2-ME system due to the stronger coordination strength of DMSO, which stabilizes lead-iodide solvent complexes and further suppresses crystallization. Consequently, both DMF- and DMSO-based binary systems yield small, discontinuous, and inhomogeneous perovskite grains with poorly defined columnar morphologies (Figs. 2c, d). In sharp contrast, the weak coordinating capacity of GVL and 2-ME endows the precursor with high crystallization reactivity, triggering instantaneous and explosive nucleation upon droplets contact the heated substrate. Such uncontrolled rapid nucleation produces densely packed yet randomly oriented perovskite grains without preferred vertical crystallographic texture (Fig. 2e).

By virtue of the rational binary solvent design, the GVL:DMSO system achieves precise modulation of precursor reactivity and solvent evaporation behavior (Supplementary Fig.3). The moderately competitive coordination of partial DMSO effectively passivates excessively reactive iodoplumbate species, thereby suppressing undesired explosive homogeneous nucleation. Meanwhile, the distinct saturated vapor pressure difference between GVL and DMSO establishes a sequential gradient evaporation mechanism during spraying. The low-boiling GVL preferentially volatilizes to induce uniform and dense nucleation at the substrate-film interface, while the residual high-boiling DMSO slowly desorbs during the subsequent growth

stage, providing sufficient temporal and spatial conditions for sustained vertical grain growth. Owing to the synergistic effects of moderated precursor activity and gradient solvent evaporation, high-quality MAPbI₃ columnar grains with uniform vertical orientation and penetrating microstructure are successfully fabricated in the GVL:DMSO system, with grain sizes exceeding 10 μm (Fig. 2f). The XRD pattern further verifies the superior crystallinity and preferred orientation of the optimized perovskite films. Specifically, the GVL:DMSO-derived MAPbI₃ film exhibits markedly enhanced diffraction intensities for the dominant (110) and (220) lattice planes, while the secondary diffraction peaks assigned to (211), (202), and (310) facets are significantly suppressed (Fig. 2g), confirming the highly oriented grain growth and improved crystalline quality.

Benefiting from the finely tuned nucleation and growth kinetics, the GVL:DMSO formulation also exhibits outstanding large-area manufacturing capability, enabling the fabrication of uniform MAPbI₃ films up to 10 × 10 cm² (Supplementary Fig.4). More importantly, the well-defined vertical columnar microstructure is perfectly retained when the spray-coating process is applied directly onto TFT substrates (Fig. 2h). The vertically aligned columnar grains form intimate, void-free interfacial contact with each individual pixel (Fig. 2i). Such superior structural integrity and favorable interfacial characteristics guarantee efficient charge extraction and uniform electrical signal readout, laying a solid foundation for subsequent high-resolution imaging applications.

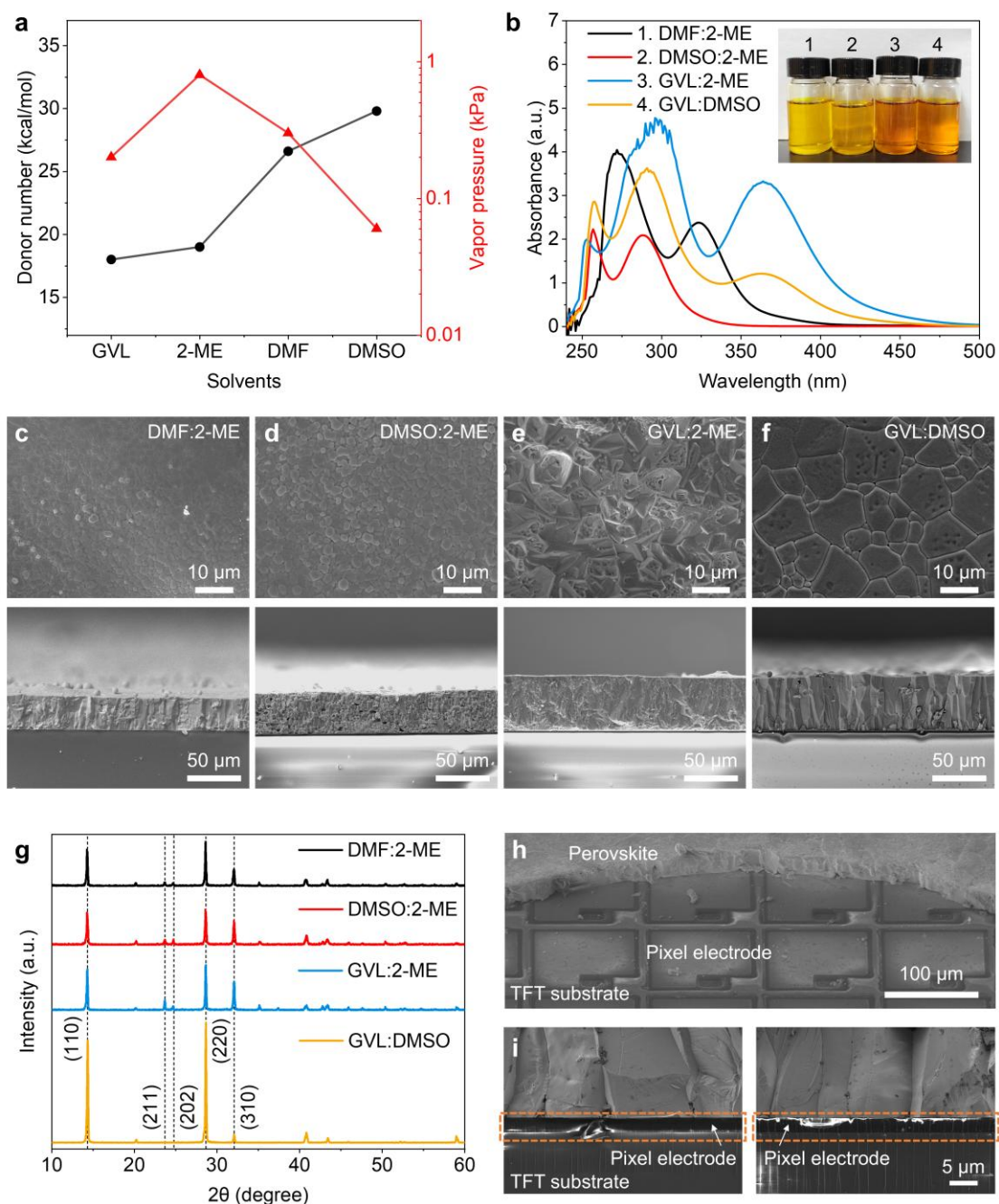


Fig. 2 | Solvent engineering for the fabrication of vertically structured bidirectional locking layer. **a**, Donor number (D_N) and vapor pressure of the four studied solvents. **b**, Ultraviolet-visible (UV-vis) absorption spectra of MAPbI₃ precursor solutions prepared from different solvent formulations. The inset shows the photo of MAPbI₃ precursor solutions with different solvent formulations at a concentration of 0.3 M. **c, d, e, f**, Surface and cross-sectional SEM images of the perovskite films prepared by solvent formulations of (c) DMF:2-ME, (d) DMSO:2-ME, (e) GVL:2-ME, and (f) GVL:DMSO. **g**, XRD spectra of the perovskite

films prepared by different solvent formulations. **h**, Integrated image of the GVL:DMSO-derived MAPbI₃ film with TFT substrate. **i**, Integrated images of MAPbI₃ columnar crystals with single TFT pixel.

The MAPbI₃ polycrystalline thick film was subsequently blade-coated on the prefabricated vertical columnar perovskite interlayer. The supersaturated nature of the perovskite precursor slurry enables only mild surface dissolution of the columnar grains, preserving the structural integrity and vertical orientation of the underlying interfacial layer. After annealing, the bilayer film was subjected to low-pressure thermo-compression treatment at 90 °C under 0.2 MPa for 1 h. This mild thermal-mechanical process efficiently eliminates intralayer voids and interfacial gaps while preventing mechanical deformation and thermal damage to the TFT substrate.

The scanning electron microscopy (SEM) was performed to elucidate the interfacial structural differences between the single-layer blade-coated film (control) and the optimized bilayer film (target). As shown in Fig. 3a and Supplementary Fig.5, the control film suffers from substantial interfacial voids and gaps at the film-substrate interface, which stem from disordered grain accumulation and inhomogeneous solvent evaporation during blade-coating. Further SEM observations of the buried interface reveal that the control film presents randomly stacked grains with abundant large pore defects (> 20 μm) at the buried interface (Fig. 3b). In stark contrast, the target film achieves seamless interfacial contact with the substrate via the bidirectional locking interlayer, with no observable delamination or void defects between the bottom columnar layer and the upper polycrystalline film (Fig. 3c) and

exhibits a dense, smooth, and defect-free buried interface (Fig. 3d). Such a compact and seamless interfacial structure favors efficient vertical carrier transport and charge collection, while simultaneously strengthening the film-substrate adhesion force.

Due to the identical MAPbI₃ composition, we speculate that the residual solvent within the blade-coated upper layer induces quasi-epitaxial growth at the bilayer interface during thermal annealing. This lattice-level coupling enables seamless grain bridging between the underlying columnar interlayer and the upper polycrystalline film, fundamentally reinforcing interfacial locking strength and guaranteeing unobstructed vertical charge transport. Such structurally compatible grain interconnection constitutes the core of our homogeneous bidirectional locking mechanism, enabling intimate interlayer integration that surpasses conventional physical contact. To further resolve the interfacial lattice configuration, focused ion beam (FIB) milling was attempted to prepare cross-sectional specimens for transmission electron microscopy (TEM) characterization (Supplementary Fig.6). However, high-energy ion beam irradiation inevitably caused interfacial delamination during sample preparation, hindering direct lattice-level TEM observation of the intact interface. Despite this limitation, SEM morphology distinctly confirms tightly interconnected grain structures across the bilayer boundary without obvious voids or stratified interfaces (Fig. 3e), solidly verifying the effective formation of the lattice-level locked bilayer architecture. This robust interlayer integration not only optimizes vertical carrier transport but also provides a favorable structural foundation for superior mechanical interfacial stability.

Micro-scratch tests were conducted to quantitatively evaluate the interfacial adhesion performance. The target film exhibits an average critical load (L_c) of 5.45 N across different test positions, which was higher than that of the control film (3.13 N), confirming enhanced interfacial tangential adhesion (Fig. 3f). Vertical tensile tests were further performed to assess the interfacial bonding strength. As depicted in Fig. 3g and Supplementary Fig.7, the target film delivers a superior tensile strength of 3.26 MPa, which is significantly higher than that of the control film (2.03 MPa). These improved interfacial mechanical properties strongly demonstrate that the homogeneous bidirectional locking strategy effectively eliminates interfacial voids and delamination, realizing a tight interconnection between the bilayer perovskite films and the underlying substrate.

Thermal shock cycling tests were further implemented to evaluate the thermal stress resistance and structural stability of the perovskite thick films (Fig. 3h). Under thermal cycling between 100 °C to 0 °C, the control film generates obvious cracks after four cycles and undergoes complete structural collapse after seven cycles (Supplementary Fig.8a). In contrast, the target film maintains excellent structural integrity even after ten cycles under the same temperature regime (Supplementary Fig.8b). Structural failure of the target film only occurs under more aggressive conditions: after three cycles between 120 °C and 0 °C, and after two cycles between 150 °C and 0 °C. The highly ordered vertical grain arrangement and reduced grain boundaries of the columnar interlayer effectively relieve internal thermal stress during

rapid temperature fluctuations, thereby endowing the target film with substantially improved structural and thermal stability.

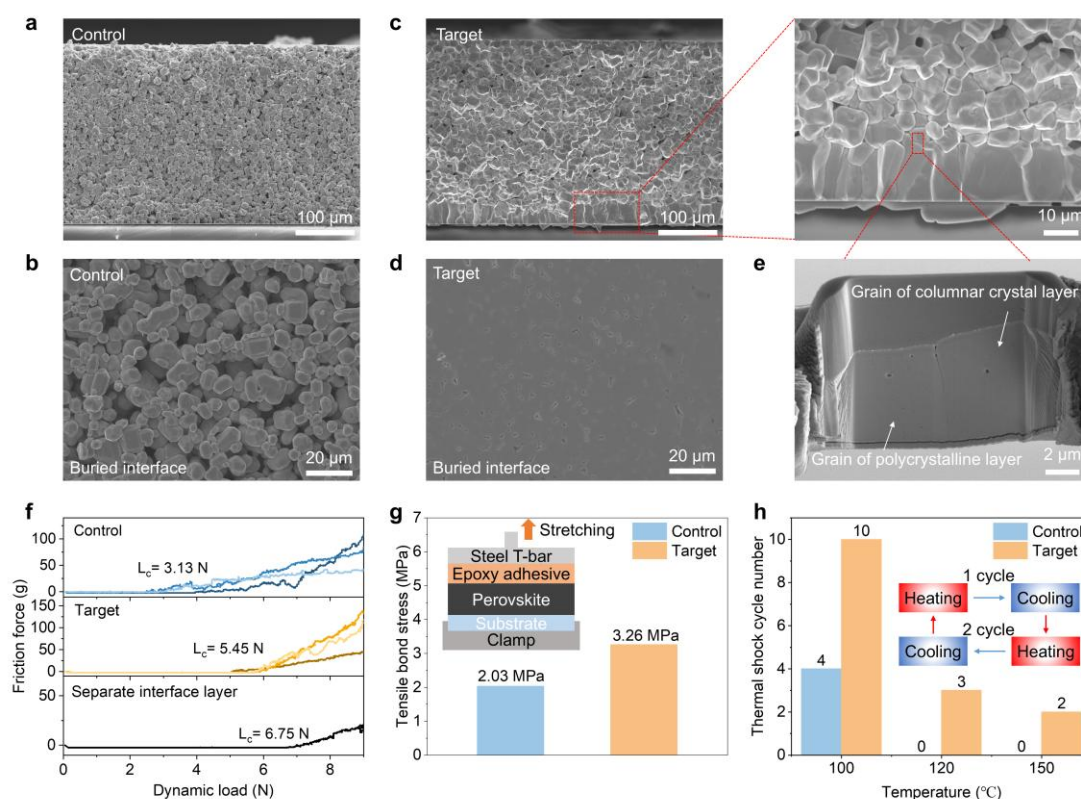


Fig. 3 | Morphology of the buried interface and mechanical properties of the devices. **a**, Cross-sectional SEM image of the control perovskite film solely prepared by blade-coating. **b**, SEM image of the buried interface of the control perovskite film. **c**, Cross-sectional SEM image (left) of the target film with a columnar crystal perovskite interfacial layer. The high-magnification SEM image (right) shows the grain contact between the blade-coating layer and the columnar crystal layer. **d**, SEM image of the buried interface of the target perovskite film. **e**, The contact of grains derived from polycrystalline and columnar crystal layers. The sample was obtained by FIB cutting. **f**, Micro scratch experiments on the control, target, and the separate interfacial films. **g**, The tensile bonding stress between the control and the target films with the substrates. The illustration shows the schematic diagram of the stretching device. **h**, The thermal shock cycle number of the control and the target devices withstanding at different temperatures.

To systematically evaluate the effects of the bidirectional-locking strategy on detection performance, vertical X-ray detectors were fabricated with indium tin oxide (ITO) serving as the bottom electrode and chromium as the top electrode. The corresponding energy band diagram of the device is displayed in Supplementary Fig.9. This simple vertical device configuration facilitates unambiguous analysis of how the buried interface layer modulates the overall detector performance.

Fig. 4a compares the dark current-voltage (I-V) characteristics of the control and the target devices. Both devices exhibit evident rectifying behavior, which originates from Schottky contact barriers formed at the metal/perovskite interfaces³⁶. Notably, the target device delivers a dark current nearly one order of magnitude lower than that of the control device, demonstrating that the engineered buried interface effectively suppresses leakage current. X-ray photoresponse measurements were further performed under a fixed electric field of -20 V mm^{-1} with varied X-ray dose rates (Fig. 4b). At a high dose rate of $83.1 \text{ } \mu\text{Gy}_{\text{air}} \text{ s}^{-1}$, the current of the target device increases from approximately -5 nA to -315 nA , yielding a high on/off ratio of 63. In contrast, the control device only achieves an on/off ratio of 16, with the current varying from -17 nA to -271 nA . Even at a low dose rate of $8.8 \text{ } \mu\text{Gy}_{\text{air}} \text{ s}^{-1}$, the target device maintains a decent on/off ratio of 8.4, whereas the control device merely retains a ratio of only 3. The significantly enhanced on/off switching behavior demonstrates that the vertically oriented columnar interlayer effectively promotes photogenerated

charge extraction and improves signal contrast under both high and low X-ray irradiation levels.

To unravel the influence of the interfacial layer on carrier transport dynamics, space-charge limited current (SCLC) measurements based on hole-only devices were carried out and the corresponding SCLC curves are displayed in Supplementary Fig.10. Both devices exhibit classical Ohmic conduction behavior at low bias voltages, where the current is dominated by thermally excited carriers with a slope of $n=1$. As the applied voltage increases, the density of injected carriers far exceeds the intrinsic carrier concentration, gradually filling the trap states and inducing a sharp current increase corresponding to the trap-filled limit (TFL) region ($n>3$). Under high applied voltages, massive injected carriers accumulate inside the perovskite film and generate a space-charge shielding effect, which restricts further carrier injection and results in a quadratic voltage dependence of the current ($n=2$). The mobility is extracted from the quadratic regime of the JV curve by fitting the Mott–Gurney equation:

$$J = \frac{9}{8} \varepsilon \varepsilon_0 \mu \frac{V^2}{L^3} \quad (1)$$

where J is the current density, V is the applied voltage, ε is the dielectric constant, ε_0 is the vacuum permittivity, L is the thickness, and μ is the mobility. The trap-filled limit voltage (V_{TFL}) of the control device is extracted to be 14.4 V, and the mobility is $6.2 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. Strikingly, the SCLC curve of the target device transitions directly from the Ohmic regime to the quadratic SCLC regime without any intermediate region, featuring a slope larger than 3. This distinct behavior stems from the drastically reduced trap density and elevated mobile carrier concentration introduced by the

oriented interfacial layer. All trap sites are fully filled under low bias, leaving no adequate voltage window to develop a prominent TFL plateau with $n > 3$ ³⁷. The target device delivers a lower V_{TFL} of 9.2 V and a remarkably enhanced hole mobility of $13.5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$. These observations verify that the homogeneous bidirectional locking interface effectively passivates both bulk and interfacial defects and facilitates long-range carrier transport across the thick perovskite absorber.

The mobility-lifetime ($\mu\tau$) product, a critical figure-of-merit for X-ray absorbers, was then extracted by fitting saturated photocurrent curves with the modified Hecht model. Benefiting from interfacial optimization, the target device achieves a prominent $\mu\tau$ product of $1.9 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1}$, which exceeds the value of $7.8 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1}$ obtained for the control device (Fig. 4c). Superior carrier transport properties facilitate efficient extraction of photogenerated charges toward electrodes, thereby improving charge-collection efficiency (CCE). The CCE under various applied electric fields is quantified via the Hecht equation³⁸:

$$CCE = \frac{L_d}{d} (1 - \exp(-\frac{d}{L_d})) \quad (2)$$

where d is the charge transport distance (equal to the device thickness), L_d is the charge drift length (equal to the product of $\mu\tau E$), and E is the electric field. As presented in Fig. 4d, with an identical film thickness of 300 μm , the target device delivers superior CCE values across all measured electric fields owing to the optimized buried interface. Importantly, a high CCE of 96.2% is achieved at a relatively low electric field of -20 V mm^{-1} , which largely circumvents stability deterioration of perovskite absorbers triggered by high bias voltages.

X-ray sensitivity is a standard metric to quantify the detection performance of radiation detectors. Fig. 4e and Supplementary Fig.11 illustrate the current density-dose rate dependence of the control and target devices under various electric fields, where the detection sensitivity is quantified by the slope of linearly fitted calibration curves. The target device retains reliable linear photoresponse across all tested electric fields and achieves an outstanding sensitivity of $6.53 \times 10^4 \mu\text{C Gy}_{\text{air}}^{-1} \cdot \text{cm}^{-2}$ at -50 V mm^{-1} , exceeding the control device ($6.31 \times 10^4 \mu\text{C Gy}_{\text{air}}^{-1} \cdot \text{cm}^{-2}$). Notably, photoconductive gain contributes to response signal amplification, and this effect is more prominent in the control device with abundant trap defects. Such gain enhancement partially offsets the performance disparity, narrowing the apparent sensitivity gap between the two devices³⁹. For this reason, we adopted the detection efficiency (DE) to further evaluate the detector performance, which combine the X-ray attenuation efficiency (α), film thickness (d), and CCE, $DE = (1 - e^{-\alpha d}) \times CCE$. Fig. 4f and Supplementary Fig.12 presents the calculated thickness-dependent X-ray attenuation ratio and DE values. Under 50 keV X-ray irradiation with an identical absorber thickness of 300 μm , the target device realizes a high DE of 69.1%. Furthermore, the DE of the target device does not undergo severe degradation upon further increasing the absorber thickness. In comparison, the DE of the control device declines rapidly, originating from restricted carrier transport. Its inferior carrier mobility leads to a carrier drift length much shorter than the absorber thickness, limiting efficient charge harvesting. High sensitivity and DE enable the target detectors maintain a high signal-to-noise ratio (SNR) under low X-ray doses. A

distinct and stable photoresponse can still be recorded at a low dose rate of $67.2 \text{ nGy}_{\text{air}} \text{ s}^{-1}$ with an SNR of 22 (Fig. 4g). By extrapolating the calibration curve at an SNR threshold of 3, the target detector achieves a low limit of detection (LoD) of $12 \text{ nGy}_{\text{air}} \text{ s}^{-1}$, outperforming most state-of-the-art polycrystalline perovskite X-ray detectors reported to date⁴⁰⁻⁴³.

Apart from the boosted photoelectric response, the homogeneous bidirectional locking buried interface also significantly improves the spatial response uniformity of the thick-film detectors. Dark and photocurrent I-V characteristics were systematically measured at nine evenly distributed electrode positions to evaluate device uniformity. In contrast to the control detector with obvious performance fluctuations, the target device exhibits negligible deviations among different testing regions, demonstrating greatly suppressed inhomogeneous response (Supplementary Fig.13). Quantitative analysis based on the relative standard deviation (RSD) of dark current and sensitivity further verifies the uniformity improvement (Fig. 4h). Operated at -20 V mm^{-1} , the control device presents poor uniformity with a dark current RSD of 22.4% and a sensitivity RSD of 9.6%. By comparison, the target device achieves remarkably reduced RSD values of only 4.7% for dark current and 1.7% for sensitivity. Long-term radiation stability was further characterized to verify operational reliability. After continuous operation for over 3 h under a cumulative X-ray irradiation dose of $100 \text{ mGy}_{\text{air}}$, the photocurrent slightly drifts from -31.9 nA to -33.2 nA , yielding a low current drift rate of $0.57 \text{ fA s}^{-1} \text{ cm}^{-1} \text{ V}^{-1}$ (Fig. 4i). Such excellent response consistency and radiation stability stem from the unique interfacial architecture

constructed via homogeneous bidirectional locking. The dense, seamless buried interface eliminates interfacial voids and weak bonding sites, which are highly susceptible to high-energy X-ray irradiation and readily evolve into detrimental deep-level defects in conventional polycrystalline perovskite films.

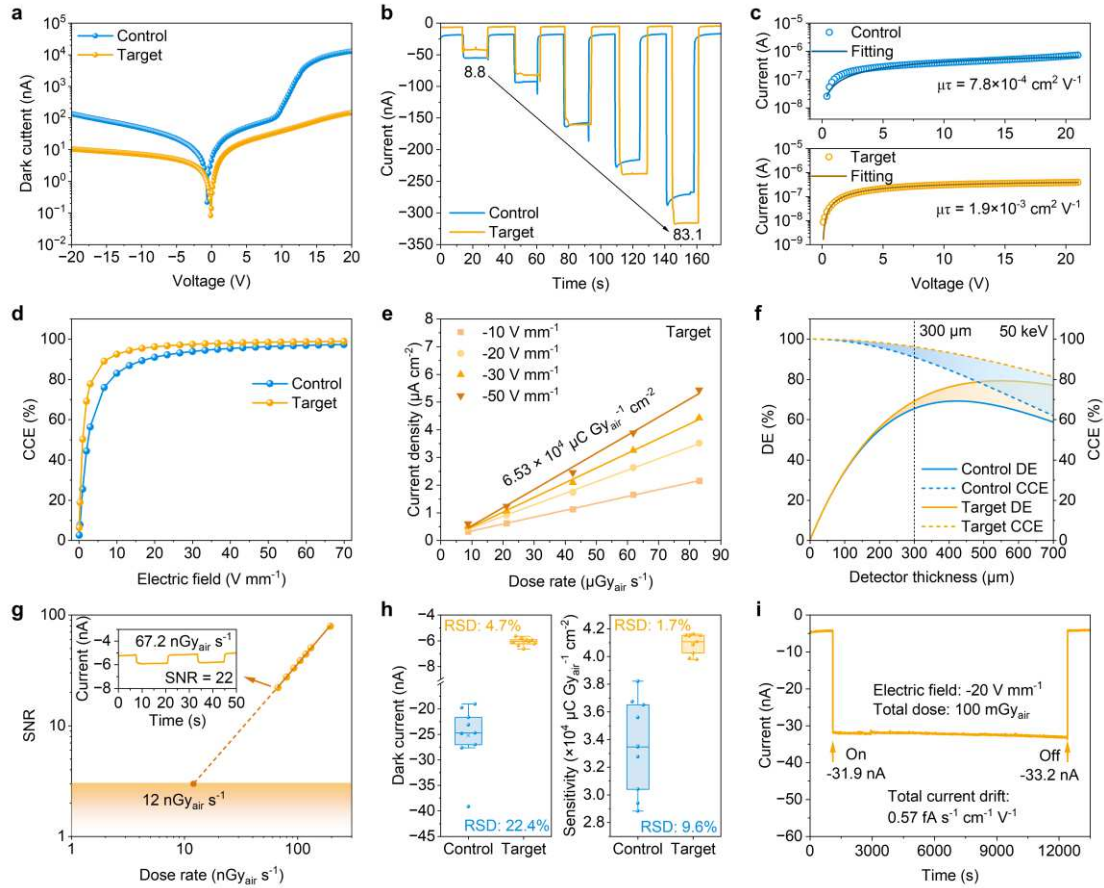


Fig. 4 | X-ray detection performances of the perovskite detectors. **a**, The dark current-voltage curves of the control and target devices. **b**, Photocurrent responses of the control and target detectors measured over the X-ray dose rates ranging from 8.8 to 83.1 $\mu\text{Gy}_{\text{air}} \text{s}^{-1}$. The applied electric field was -20 V mm $^{-1}$. **c**, Bias-dependent photocurrent of the control and target devices. **d**, Bias-dependent charge collection efficiency of the control and target devices. **e**, The linear relationship of photocurrent density with X-ray dose rate under different electric fields. **f**, Thickness-dependent DE and CCE of the control and target detectors for 50 keV X-ray. **g**, Signal-to-noise ratio of the target detector under different dose rates. The inset shows the photocurrent response at a dose rate of 67.2 $\text{nGy}_{\text{air}} \text{s}^{-1}$. **h**, RSD of the dark current and the

sensitivity of the control and target detectors. **i**, The X-ray radiation stability of the target detector.

The comprehensive improvements in interfacial integration and X-ray detection performance motivate us to further integrate the optimized bilayer perovskite thick film with TFT arrays for the fabrication of X-ray imaging panels. Fig. 5a presents the assembled perovskite X-ray imaging system integrated with gate drive integrated circuits (ICs) and flexible printed circuits (FPCs). The fabricated imaging panel possesses a 256×256 -pixel array, an active area of $22 \text{ mm} \times 22 \text{ mm}$, and a pixel pitch of $85 \text{ }\mu\text{m}$, enabling high-resolution spatial imaging acquisition.

Flat-field imaging tests were performed under dark conditions and uniform X-ray irradiation to compare the imaging homogeneity of control and target detectors (Fig. 5b). The target imaging panel exhibits exceptional spatial uniformity with negligible dead pixels of 0.28% (Supplementary Note 1) and uniform gray-scale distribution across the entire detection area. In contrast, the control detector suffers from a severely inhomogeneous response, accompanied by widespread gray-scale fluctuation and numerous defective pixels, with a dead-pixel rate as high as 1.13%. Three-dimensional gray-value distribution mapping further intuitively verifies the optimized imaging uniformity of the target device (Fig. 5c). Both dark-field and bright-field images reveal an extremely smooth and consistent pixel gray-scale distribution for the target detector, while the control device displays prominent spatial fluctuations.

For pixelated X-ray image sensors, pixel-to-pixel response inconsistency deteriorates the signal-to-noise ratio, introduces contrast distortion, and ultimately degrades the spatial resolution of resulting images. To quantitatively evaluate the response uniformity, we statistically analyzed the response current of all 256×256 pixels under X-ray irradiation. The pixel intensity distribution was fitted with a Gaussian function, and the photo response non-uniformity (PRNU) was defined as the ratio of the standard deviation to the mean value of all pixel responses (Fig. 5d). The control device exhibits a high PRNU of 28.57%, whereas the target device achieves a remarkably low PRNU of only 4.31%. This dramatic suppression of pixel response variation confirms that the seamless, defect-free buried interface effectively homogenizes interfacial contact and carrier extraction across the TFT array, eliminating localized charge-collection dead zones and greatly improving the imaging consistency of the thick-film perovskite detector.

The spatial resolution of the X-ray imaging detectors was further evaluated via the modulation transfer function (MTF), which was calculated with the slanted-edge method (Fig. 5e). The MTF curve quantitatively characterizes the detectors' capability to reproduce fine spatial details. At a threshold MTF value of 0.2, the control detector delivers a spatial resolution of 4.8 lp mm^{-1} , while the interface-optimized target detector achieves an enhanced resolution of 6.1 lp mm^{-1} . The superior resolution of the target device is further validated by imaging standard line-pair test patterns. The control detector resolves a maximum spatial frequency of only 4.6 lp mm^{-1} (Fig. 5f), whereas the target detector successfully distinguishes fine line structures up to 6.0 lp

mm⁻¹ (Fig. 5g). Such a prominent spatial resolution enhancement originates from the vertically oriented columnar crystalline architecture and compact interfacial integration, which effectively suppresses lateral carrier diffusion and inter-pixel crosstalk, thereby enabling the preservation of high-fidelity spatial information during X-ray photoelectric signal conversion and readout.

Considering the superior pixel uniformity and improved spatial resolution, the optimized target perovskite X-ray detector exhibits promising applicability for practical non-destructive testing and low-dose biomedical imaging. Fig. 5h presents the X-ray imaging result of a commercial chip, in which delicate pin architectures and tiny damaged defects can be clearly identified, demonstrating the excellent fine-feature recognition capability of the optimized imaging panel for industrial non-destructive scenarios. Furthermore, low-dose biological imaging was performed at a low radiation dose of 4.72 $\mu\text{Gy}_{\text{air}}$ per frame. As depicted in Fig. 5i, the skeletal framework and soft tissue contours of a dried fish are distinctly resolved, validating the great potential of our perovskite detector for low-dose biomedical radiographic imaging.

Beyond conventional two-dimensional planar imaging, we further explored the feasibility of the optimized detector for X-ray computed tomography (CT) reconstruction. The schematic configuration of the CT imaging system is shown in Fig. 5j. A spring encapsulated within a capsule was selected as the test sample and rotated with an angular step of 2°, yielding a total of 180 projection images at diverse viewing angles for tomographic. The single-projection X-ray image clearly resolves

the outline of the capsule and the internal spring structure (Fig. 5k). The reconstructed two-dimensional tomographic slices along the XY and YZ planes accurately visualize the internal assembly morphology of the sample (Fig. 5l). Furthermore, complete three-dimensional reconstruction successfully visualizes the overall morphology of the inner spring and outer capsule (Fig. 5m). These comprehensive imaging demonstrations solidly verify that the interface-modified perovskite X-ray detector possesses outstanding imaging fidelity and exquisite structural detail distinguishability, enabling high-precision 2D planar imaging and 3D reconstruction for versatile X-ray imaging applications. A comprehensive performance benchmark between our detector and previously reported pixelated direct-conversion perovskite X-ray imagers is summarized in Supplementary Table 1. The comparative results further verify the outstanding comprehensive performance of our bilayer device and demonstrate the superiority of the proposed buried interface engineering strategy toward high-quality X-ray flat-panel imaging.

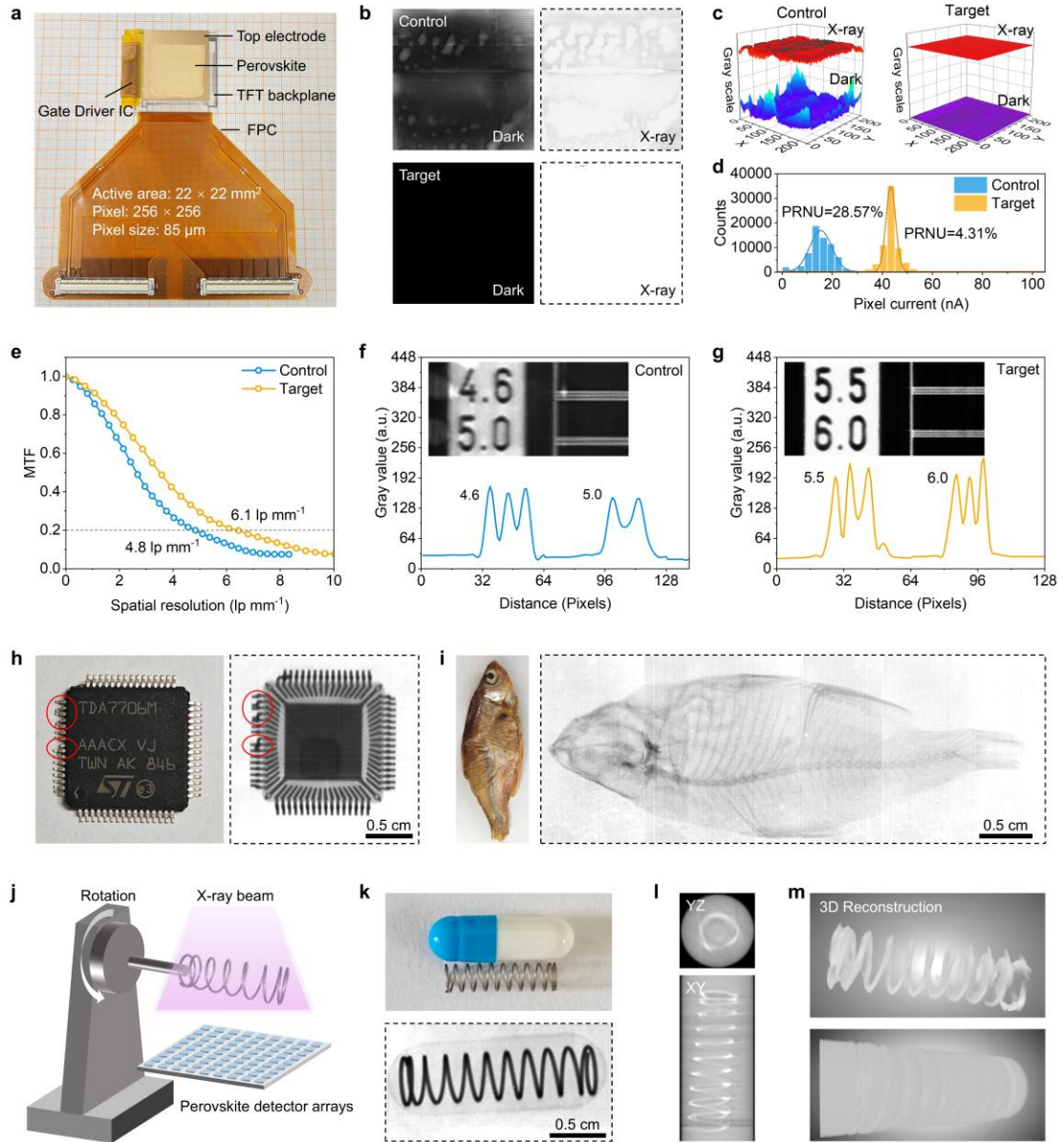


Fig. 5 | X-ray imaging performance and application demonstrations. **a**, Photograph of the fabricated perovskite X-ray imaging panel integrated with a TFT array, gate driving IC, and FPC. **b**, The acquired images of the control and target detectors under darkness and X-ray exposure. **c**, 3D pixel gray-scale distribution of the control and target detectors. **d**, Statistical distribution and PRNU analysis of pixel photoresponse current of the control and target detectors. **e**, MTF curves of the control and target detectors. **f**, **g**, X-ray imaging and greyscale fitting results of a standard resolution card of the control (**f**) and target (**g**) detectors. **h**, X-ray nondestructive imaging of a commercial chip with fine pin defects. **i**, X-ray imaging of a fish specimen at a dose rate of $4.72 \mu\text{Gy}_{\text{air}}$ per frame. **j**, Schematic illustration of the X-ray

CT equipment. **k**, Single-angle X-ray projection of a spring-encapsulated capsule sample. **l**, Reconstructed 2D tomographic slices in XY and YZ planes. **m**, 3D reconstructed CT image of the encapsulated spring structure.

Conclusion

In conclusion, this work develops a distinctive homogeneous bidirectional locking interfacial engineering strategy to address the critical bottlenecks of solution-processed thick perovskite X-ray flat-panel detectors, including the thickness limitation, thermal expansion mismatch, defective and weakly bonded buried interface. Utilizing a composition-consistent vertically oriented columnar perovskite interlayer, the proposed structure achieves robust bidirectional interfacial locking: the bottom columnar layer tightly bonds with TFT pixel electrodes to eliminate contact dead zones and alleviate thermal expansion mismatch, while the top interface enables quasi-epitaxial growth and seamless fusion with the overlying blade-coated polycrystalline absorber, effectively eliminating interfacial voids and structural delamination. Benefiting from this unique bidirectional locking effect and continuous vertical charge transport channels, the optimized device exhibits comprehensively enhanced performance. Specifically, the interfacial tensile strength is improved by 61% with enhanced thermal shock resistance, the photoresponse non-uniformity is remarkably reduced from 28.57% to 4.31% with dead-pixel rate of 0.28%, and a competitive spatial resolution of 6.1 lp mm⁻¹ is achieved, enabling high-fidelity 2D radiography and reliable 3D CT reconstruction. This work breaks the trade-off between thick-film X-ray attenuation capability, interfacial integrity, device mechanical robustness and imaging uniformity

of polycrystalline perovskite X-ray detectors, offering a facile and scalable pathway toward the development and industrialization of high-resolution, low-dose perovskite X-ray flat-panel imaging systems.

Methods

Materials

Lead iodide (PbI_2 , 99.9%) was purchased from Shanghai Macklin Biochemical Co., Ltd. Methylammonium iodide (MAI, 99.5%) was purchased from Xi'an Yuri Solar Co., Ltd. DMF, DMSO, 2-ME, and anhydrous acetonitrile (ACN, 99.9%) were purchased from Shanghai Aladdin Bio-Chem Technology Co., Ltd. GVL and was purchased from Tianjin Xiensi Biochemical Technology Co., Ltd. All the chemicals were used without further purification.

Fabrication of precursor solution and polycrystalline powder

For ultrasonic spray coating, MAPbI_3 precursor solutions were prepared by dissolving PbI_2 and MAI at a molar ratio of 1:1.1 in four different mixed solvent systems, including DMF:2-ME, DMSO:2-ME, GVL:2-ME, and GVL:DMSO with a fixed volume ratio of 4:1, to achieve a uniform concentration of 0.3 mol L^{-1} . For the preparation of MAPbI_3 microcrystalline powder, PbI_2 and MAI were mixed at a molar ratio of 1:1.4 and dispersed in anhydrous ACN. The mixture was continuously stirred at 60°C for 6 h to fully generate black perovskite MAPbI_3 precipitates. The resulting suspension was centrifuged at 8000 rpm for 2 min to collect the precipitates, followed by repeated centrifugal washing with anhydrous ACN three times. The purified black precipitates were dried overnight in a vacuum oven at 60°C and finally ground into uniform MAPbI_3 microcrystalline powder for subsequent use.

Fabrication of bilayer perovskite films

Commercial ITO-coated glass substrates and TFT array substrates were ultrasonically cleaned sequentially with deionized water, ethanol, and isopropanol for 15 min each, followed by nitrogen blow-drying and ultraviolet-ozone treatment for 20 min before film deposition. The vertically columnar perovskite interlayer was prepared via an ultrasonic spray-coating strategy. The MAPbI_3 precursor solution was delivered to the spray nozzle at a feeding rate of $300 \mu\text{L min}^{-1}$ using a stepping-motor-driven syringe. Under ultrasonic atomization, the precursor solution was transformed into uniform aerosol droplets and deposited onto the preheated substrate under a spraying pressure of 0.016 MPa, with the substrate temperature maintained constantly at 120°C . The nozzle scanned along the X-axis at a moving speed of 80 mm s^{-1} with an inter-line step of 3 mm for 20 scanning cycles, followed by another 20 repeated scanning cycles along the Y-axis with identical scanning parameters. A total of 100 dual-axis scanning cycles were performed to complete the deposition of the columnar perovskite seed layer. After spray coating, the obtained film was thermally annealed for 1 h and subsequently cooled slowly to room temperature at a controlled cooling rate of approximately 1°C min^{-1} to gradually release residual internal stress. Subsequently, a thick perovskite absorber layer was blade-coated on the spray-deposited columnar interlayer to construct the bilayer architecture. A highly supersaturated perovskite slurry (10 mol L^{-1}) was prepared by dispersing the pre-synthesized MAPbI_3 microcrystalline powder in GVL solvent. The gap height between the blade and substrate was fixed at $400 \mu\text{m}$ during blade coating. The as-prepared bilayer wet film

was annealed at 90 °C for 3 h to complete grain crystallization and film formation. After annealing, the bilayer film was further treated via low-temperature and low-pressure thermal compression at 90 °C under 0.2 MPa for 1 h. This mild post-treatment effectively eliminates interlayer voids and interfacial gaps without introducing mechanical or thermal damage to the underlying substrate, thereby realizing dense, seamless, and tightly bonded bilayer perovskite thick films.

Perovskite Film Characterization

The absorption spectra of perovskite precursor solutions with different solvent formulations were measured using a UV-Vis-NIR spectrophotometer (Shimadzu UV-3600i Plus). All precursor solutions were diluted 1000-fold prior to spectroscopic testing to satisfy the measurement concentration range. XRD patterns of the columnar perovskite films were acquired using a Bruker-AXS D8 Advance diffractometer to analyze crystalline structural features and orientation. The surface, cross-sectional, and buried interfacial morphologies of the perovskite thick films were observed via field emission SEM (JSM-IT500HR) with an accelerating voltage of 15 kV. A FIB-SEM dual-beam system (FEI Scios HiVac) was employed for precise FIB cutting and in-situ morphological observation of the grain-contact region between the columnar interlayer and the upper polycrystalline film. The interfacial adhesion strength between perovskite films and substrates was quantitatively evaluated through two mechanical testing methods. Scratch tests were performed using a micro-scratch tester with an applied load of 10 N. Furthermore, a tensile bonding test was conducted

to measure the film-substrate interfacial tensile strength. The glass substrate was rigidly fixed by a fixture, while the film surface was bonded to a steel T-bar via epoxy resin. The tensile stress was recorded in real time using an adhesion tester (Positest AT-M) to determine the interfacial bonding force. For thermal shock cycling evaluation, the perovskite film was placed on a preheated substrate for 1 min and immediately transferred to a pre-cooled substrate for another 1 min, which was defined as one complete thermal shock cycle.

X-ray detection performance characterizations

X-ray detection measurements were performed using a silver-anode X-ray tube operated at a fixed acceleration voltage of 50 kV in accordance with the RQA3 standard. The X-ray dose rate was precisely tuned by adjusting the tube current ranging from 10 to 80 μ A and altering the distance between the X-ray source and the detector. A 5 mm-thick aluminum sheet was placed between the source and the device as an X-ray attenuator, and the actual dose rate was calibrated using an ionization chamber scanner (Fluke 481). The I-V and I-t response curves of perovskite detectors were synchronously recorded by a Keithley 2400 source meter. All X-ray detection and imaging experiments were carried out at room temperature inside a lead shielding box to eliminate external radiation interference.

X-ray imaging and CT reconstruction

For X-ray imaging characterization, the bilayer perovskite active layer was directly

fabricated on the active area of a TFT pixel array substrate, and gold electrodes were thermally deposited on the top of the perovskite layer and common electrode region without a shadow mask. The pin area of the TFT substrate was electrically connected to the signal readout system via a flexible printed circuit. The 256×256 TFT imaging chip (model: SC-T-G256-2402 (GHS2)) and the matched LinkZill 03MR readout device were provided by Hangzhou LinkZill Technology Co., Ltd., operating under gate IC driving mode. During imaging acquisition, pixel current signals were read line-by-line with a line scanning time of 0.43 ms, corresponding to a frame rate of 9 frames per second (FPS). The acquired raw signals were transmitted to computer via USB, processed by built-in imaging algorithms, and finally reconstructed into 256-level grayscale images. The spatial resolution of the imaging panel was quantitatively evaluated via the MTF based on the slanted-edge method. A 1.0 mm-thick tungsten plate was placed on the pixel array at a small tilt angle to acquire slanted-edge radiographic images. The edge spread function (ESF) was extracted through sub-pixel interpolation, followed by derivation to obtain the line spread function (LSF). The MTF curve was finally calculated via Fourier transformation of the LSF profile. X-ray CT imaging was performed on a self-built radiographic platform. The incident direction of the X-ray beam was vertically aligned with the scanned object and the detector. The distance between the X-ray source and the scanned object was fixed at 200 mm, and the distance between the X-ray source and the detector was set to 250.0 mm. During CT scanning, the target object was mounted on a precision rotary stage and scanned over a full 360° rotation with an angular step

of 2°, yielding a total of 180 projection images at different viewing angles. All acquired planar projection images were imported into Dragonfly software for post-processing and three-dimensional tomographic reconstruction.

Data and materials availability

Source data are provided with this paper. Other data that support the findings of this study are available from the corresponding authors upon reasonable request.

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Author Contributions

X.L. and C.Z. supervise this project. Y.C. was responsible for all the experiments in this research, including material preparation, device fabrication, optical, mechanical and electronic characterization, as well as X-ray imaging. Y.R. assisted in X-ray

image processing and CT imaging. X.J. and J.L. assisted in mechanical testing. Z.W., Y.G., and A.L. assisted in material preparation. Y.C. and X.L. prepared the manuscript. X.L., H.Z., and C.Z. made revisions to the manuscript. All authors discussed the results and confirmed the manuscript.

Competing of Interests

The authors declare that they have no competing interests.

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