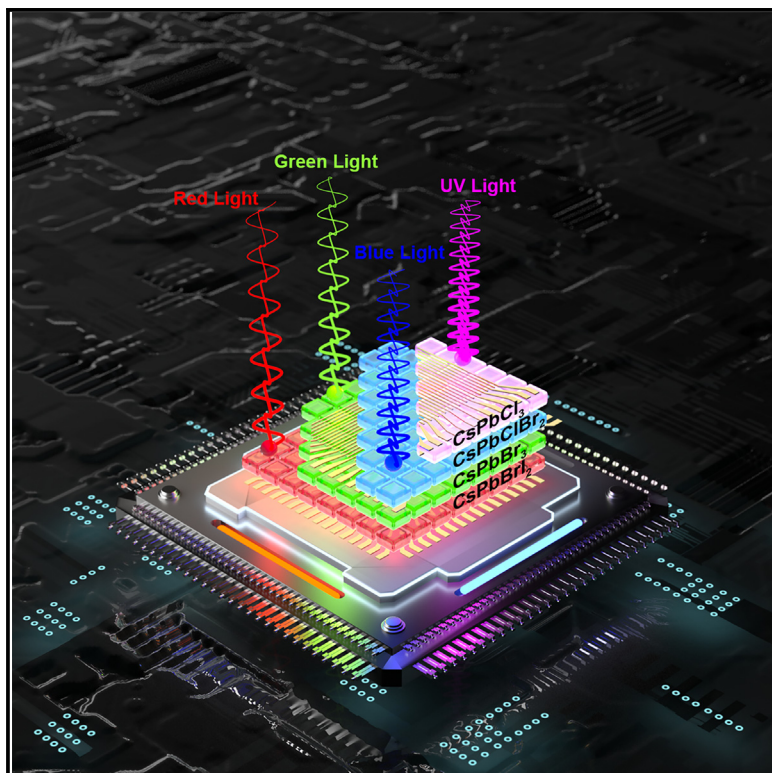


A tetrachromatic sensor for imaging beyond the visible spectrum in harsh conditions

Graphical abstract



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In brief

The fully integrated tandem all-inorganic perovskite imaging system can detect ultraviolet, blue, green, and red light simultaneously. With four 32×32 color-selective pixel arrays, the device demonstrates reliable performance in extreme temperatures and atmospheric conditions, including aggressive thermal and vacuum cycles. The sensor offers a compact and reliable solution for tetrachromatic imaging in harsh environments, such as for space exploration and self-navigating vehicles.

Highlights

- An all-inorganic perovskite-based tandem image sensor is presented
- The tetrachromatic sensor can capture images in the ultraviolet and visible spectra
- It can operate reliably and withstand thermal cycling from 150 to 500 K



Develop

Prototype with demonstrated applications in relevant environment



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Article

A tetrachromatic sensor for imaging beyond the visible spectrum in harsh conditions

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THE BIGGER PICTURE With the increasing demand for image sensor technology, the integration of multi-spectral and visible/non-visible light sensing systems can help expand the capabilities of applications such as geographic remote sensing, biological monitoring, and space exploration. This research introduces an integrated wide-spectrum imaging system that combines ultraviolet and red-green-blue four-color channels, utilizing four layers of all-inorganic perovskites as the light-sensitive layers. The resulting sensor demonstrates exceptional reliability, operating consistently in atmospheric and vacuum environments from 150 to 500 K. This offers a compact solution for multispectral detection, particularly in challenging conditions for extravehicular operations and autonomous driving.

SUMMARY

A tetrachromatic light sensor can detect light in four independent color channels and extend its detection range beyond the visible spectrum. However, current tetrachromatic sensors face challenges such as the inability to work reliably in extreme temperatures. In this work, we present a fully integrated tandem tetrachromatic perovskite imaging system. Four layers of all-inorganic perovskites in the device serve as ultraviolet, blue, green, and red light detectors and can discriminate spectral and non-spectral colors. The tandem image sensor comprises four 32×32 color-selective pixel arrays. The non-encapsulated device is tested to function from 150 to 500 K under atmospheric pressure and vacuum environments. The fabrication process and device structure are scalable, with the potential to be used for tetrachromatic imaging under harsh environmental conditions.

INTRODUCTION

The biological world provides a diverse range of abilities within color vision.¹ In comparison to humans, some species of birds have cone receptors in their retinas that extend their visual range

to the ultraviolet (UV) spectrum to help them discriminate organisms with differences in UV absorption rates, navigate through the landscape, detect food and predators, and select their mates.^{2,3} In practical applications, tetrachromatic light sensors have usage in scenarios such as environmental monitoring,



including monitoring foliage and vegetation physiology by capturing multiple wavelengths of light and mapping for geology through the analysis of reflectance patterns at different wavelengths, as well as in industrial applications, such as implementation of multispectral lidar systems for autonomous driving, inspection of power infrastructure, and quality control in the industry.^{4,5}

Inorganic semiconductors, such as GaN, Si, PbS, and InGaAs, have so far been the predominant materials typically used in optoelectronics, as they offer absorption capabilities across a wide range from UV to infrared wavelengths. However, the complexity and cost of heterogeneous integration have created challenges when fabricating devices for sensing in the UV and red-green-blue spectrum (UV + RGB). Despite these challenges, recent research exploring alternative materials such as metal oxide, organic, and 2D materials has showcased promising advancements in this field. For instance, Li et al. presented a vanadium dioxide-based UV neuromorphic UV sensor for image optimization,⁶ and Wu et al. demonstrated a tetrachromatic optical synaptic device using a 2D tungsten diselenide optoelectronic p-type transistor for fully light-controlled bidirectional synaptic excitation and inhibition, emulating reindeer behavior.⁷ Jiang et al. reported a controllable UV-ultrasensitive neuromorphic vision sensor based on organic phototransistors with excellent optical figures of merit, enhancing recognition accuracy in UV image recognition.⁸ However, these works primarily focused on single-active-layer broadband image sensors and lacked the ability to accurately discriminate different color signals and reconstruct images with four channels.

Perovskite-based photodetectors have recently emerged as promising alternatives to these image sensors, with features such as high responsivity, high detectivity, fast response time, wide-dynamic-range tunable band gap, and low fabrication costs.^{5,9,10} Particularly, inorganic halide perovskites show the advantages of superior thermal stability with low-cost and efficient preparation methods. However, halide-perovskite-integrated imaging systems face challenges of solution process compatibility when more than two halide perovskite layers are stacked on each other and have poor compatibility with existing microfabrication and device miniaturization strategies, such as photolithography.^{11,12} Specifically, the existing strategies for constructing multicolor perovskite photodetectors uses either a parallel or a stacking design. Parallel structures necessitate a color filter array for monochromatic detection, which reduces efficiency, spatial resolution, and color accuracy,^{12,13} while stacking structures consist of multiterminal tandem perovskite units, thus increasing the cost and the complexity of processing, especially on pixel alignment. In this regard, integrating colorful perovskite imaging systems in one device is highly desirable.

In this work, we present a fully integrated UV + RGB all-inorganic perovskite image sensor system with four layers of 32 × 32 pixel arrays, in which CsPbX₃ perovskites serve as the photosensitive layers and metal oxides serve as the charge transport layers. The four sequentially thermal-evaporated perovskite layers exhibit high crystallinity, low defect density, and long carrier lifetime. The thickness of each layer is optimized through simulations to achieve over 99.5% color selectivity while maintaining high light transmittance. A tandem dual-band strategy

is used for the image sensor to dispense light without external filters and improve the optical efficacy.^{14–16} The integrated tetrachromatic image sensor works well under harsh environments, retaining over 95% responsivity after over 1,000 thermal cycles from 150 to 500 K and can remain functional down to 16 K.

RESULTS AND DISCUSSION

Selection and thickness optimization of the perovskite thin films

The tunable band gap of perovskite, modulated by halide ion mixing, enables four perovskite active layers: CsPbCl₃, CsPbClBr₂, CsPbBr₃, and CsPbBrI₂, which, respectively, mimic the UV- or violet-sensitive, short-wavelength-sensitive, medium-wavelength-sensitive, and long-wavelength-sensitive cones found in some species of birds. The CsPbCl₃ layer serves as a broadband UV image sensor, while the other layers function as single-color image sensors due to the color filter effect in the stacked architecture. As shown in Figure 1A, a thin quartz layer separates the device into two halves, one with the UV and blue sensors and the other with green and red sensors. To estimate the distribution of optical intensity and photon absorption for device design, we simulated the optical properties of all layers (Figure 1B). The required complex refractive indices (n, k) were obtained from UV-visible (UV-vis) spectroscopy and spectroscopic ellipsometry (Figure S1). Additionally, we performed optical simulation on the transmittance of four photoactive perovskite layers as a function of film thickness (Figure S2). Due to the sensor's stacked architecture, the lower layers require a high light intensity to achieve high responsivity, creating a trade-off between color selectivity and device performance. We conclude that the light leakage should be below 0.5% across the entire detection range to avoid crosstalk in light discrimination (the simulation results are shown in Figures S2 and S3). We chose the optimal thicknesses of 200, 300, 320, and 350 nm for the CsPbCl₃, CsPbClBr₂, CsPbBr₃, and CsPbBrI₂ thin films, respectively. Figure 1C plots the measured transmittance of specific-thickness thin films derived from UV-vis (Figure S4). The photon absorption simulation profile shown in Figure 1C reveals that photons with wavelengths ranging from 300 to 410, 410 to 485, 485 to 520, and 520 to 650 nm are primarily absorbed in the CsPbCl₃, CsPbClBr₂, CsPbBr₃, and CsPbBrI₂ layers. Indium tin oxide (ITO) electrodes, quartz, and charge-transporting layers contribute minimally to photon absorption, as substantiated by the UV-vis of these layers (Figure S4).

The perovskite layers are fabricated using a scalable physical vapor deposition process. Sequential thermal evaporation is used to achieve high-quality crystalline all-inorganic perovskite thin films with layer-by-layer process control while maintaining film uniformity, conformal substrate coverage, and compatibility with microfabrication processes.^{17,18} Figure S5 presents top-view and cross-sectional scanning electron microscope (SEM) images of the perovskite thin films and device structure, respectively. Atomic force microscopy (AFM) images in Figure S6 further verify that the obtained perovskite films are dense with a root-mean-square roughness below 20 nm. The UV-vis spectra in Figure S4 exhibit high excitonic peaks at absorption edges, which decay with the increase in absorption wavelength. This

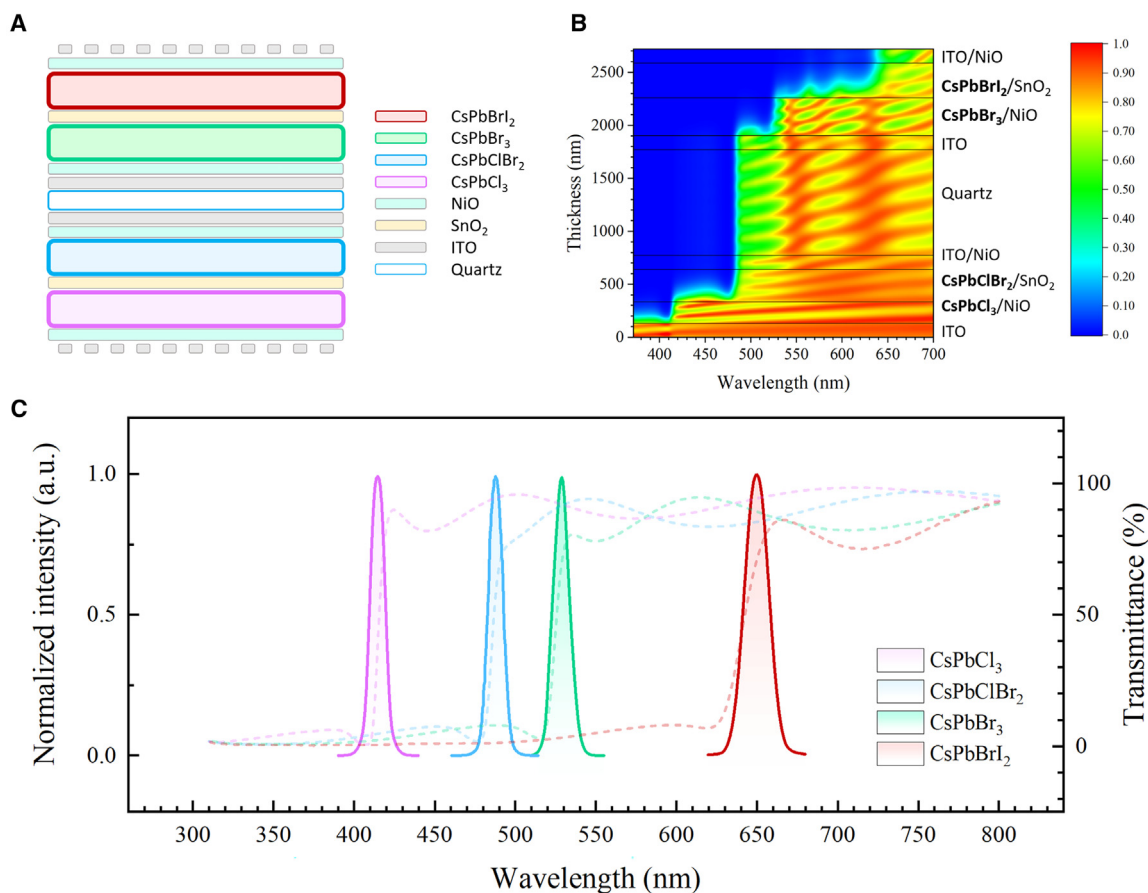


Figure 1. Overall tetrachromatic perovskite image sensor design

- (A) Schematic of tetrachromatic image sensor structure.
 (B) Simulation of the wavelength-dependent optical field with respect to device position.
 (C) UV-vis and PL spectra of four perovskite thin films.

implies that the evaporated perovskite thin films have high crystallinity, as previous studies have suggested that excitons are more likely to form at locations with higher crystallinity and promote light absorption.¹⁹

The band gaps of the four perovskite materials are 3.01, 2.57, 2.36, and 1.93 eV, as estimated by the Tauc plots shown in the inset images of Figure S4. Solid lines in Figure 1F exhibit the photoluminescence (PL) with a narrow full width at half maximum (FWHM) between 20 and 35 nm. X-ray photoelectron spectroscopy (XPS) results (Figure S7) reveal a Pb 4*f* binding energy shift suggesting a strong interaction between Pb and Cl atoms in the [PbCl₆]⁴⁻ octahedral group. X-ray diffraction (XRD) spectra shown in Figure S8 confirm the high crystallinity of perovskite thin films. Specific XRD structural refinement results by FullProf are shown in Figures S9 and S10, which agree with the diffraction peaks and standard room temperature structures and show the impact of halogen ion densities on thin film stability. Except for the active perovskite layers fabricated by thermal evaporation, the remaining layers of metal oxide electrodes and charge-transporting layers are continuously prepared via magnetron sputtering. Metal oxide charge-transporting layers prepared by sputtering are dense, as substantiated by the

AFM morphology images in Figure S11. The fabrication process is shown in more detail in Figures S12–S14.

Working mechanism and basic characterization of the tetrachromatic sensor

UV photoelectron spectroscopy (UPS) and optical band-gap measurements were used to determine the valence and conduction band energies (E_v and E_c) of the NiO hole-transporting layer and the SnO₂ electron-transporting layer (ETL), as shown in Figure S15. To discriminate among the four spectral colors, we used two p-i-n-i-p structures (as illustrated by the energy-level alignment without bias in Figure 2A): one for detecting UV and blue light and the other for detecting green and red light. Each p-i-n-i-p configuration comprises a wide-band-gap perovskite p-i-n diode and a narrow-band-gap perovskite n-i-p diode. The reason for selecting SnO₂ as the ETL material is the large valence band offsets (i.e., ΔE_{v1} , ΔE_{v2} and $\Delta E'_{v1}$, $\Delta E'_{v2}$) between the perovskite layer and the n-type semiconductor.^{14,20} These offsets act as hole barriers preventing hole injection at the perovskite-ETL interface.²¹

A positive or negative bias is applied to the two side electrodes, activating one diode and deactivating the other within

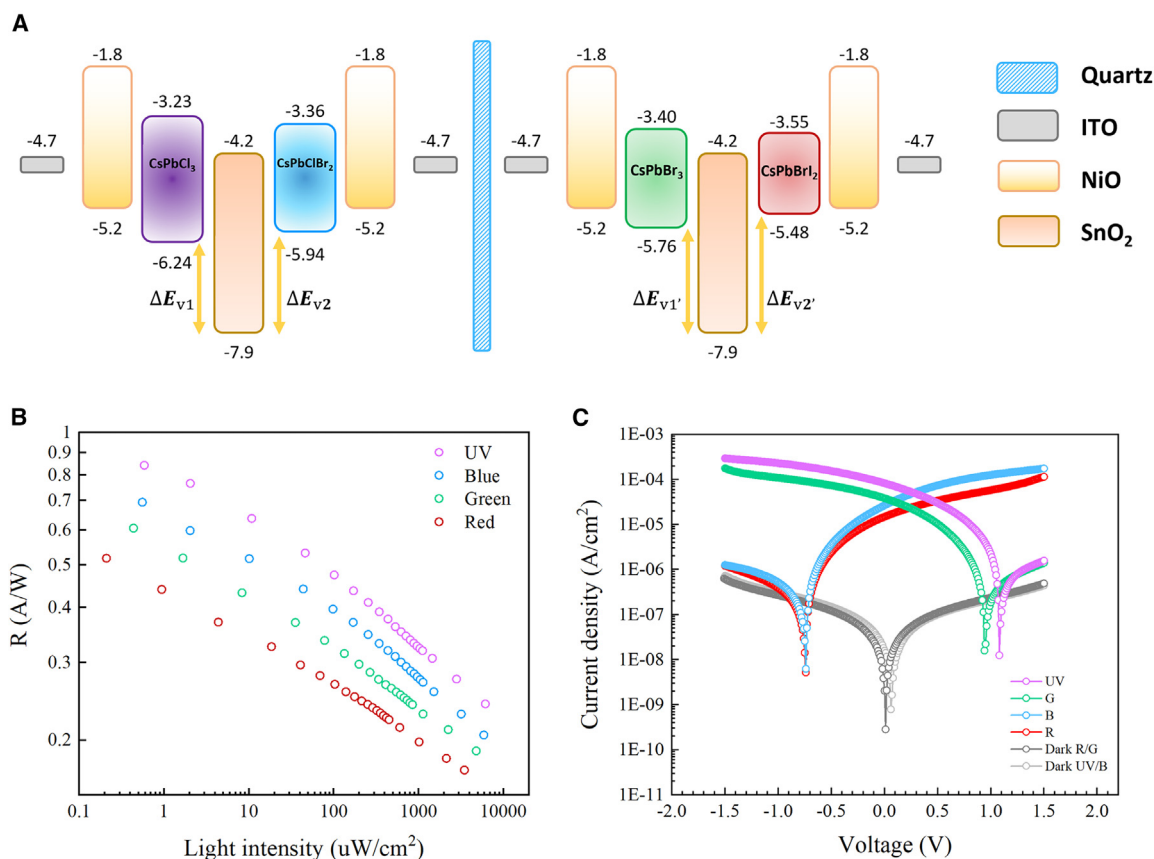


Figure 2. Photoelectric response characteristics of device

(A) Energy-level alignment of the device. The unit of the values in the energy level diagram is eV.

(B) Responsivity of the tetrachromatic image sensor as a function of incident light intensity under 405, 450, 520, and 635 nm light source.

(C) Density-to-voltage (J-V) characterizations of tetrachromatic image sensor under dark and single-wavelength illuminations.

the p-i-n-i-p configuration to distinguish short- and long-wavelength light signals. In the photodiode mode, photogenerated carriers exhibit a greater tendency to separate under reverse mode, i.e., when the positive terminal of the source is connected to the n side and the negative terminal is connected to the p side, whereas the opposite is true for the forward mode. When a positive bias is applied to the sensor, as depicted in Figure S16A, the NiO/(CsPbCl₃ or CsPbBr₃)/SnO₂ diode (p-i-n diode) is under reverse mode, while the SnO₂/(CsPbClBr₂ or CsPbBrI₂)/NiO diode (n-i-p diode) is under forward mode. The perovskite layer of the p-i-n diode serves as the photoactive layer, generating photogenerated carriers. After the electron-hole pairs separated, the electrons would drift toward the interface between the other perovskite layer and the ETL to recombine with the holes that had accumulated there due to the large valence band offset. In contrast, the n-i-p diode functions as a conduction layer, and no photocurrent is generated, even if long-wavelength light produces photogenerated carriers. These carriers cannot separate and transfer due to band bending under forward mode. Conversely, long-wavelength light is detected when a negative bias is applied, as shown in Figure S16B.

The optoelectronic properties of tetrachromatic image sensors were investigated using four single-wavelength lasers cor-

responding to UV + RGB light at 405, 450, 520, and 635 nm. As demonstrated by Figure 2B, the responsivities of the four sensors are presented as a function of a light intensity ranging from ~ 0.2 to ~ 6 mW/cm². The maximum responsivity values at 405, 450, 520, and 635 nm are 0.842, 0.694, 0.606, and 0.518 A/W under light intensities of 0.586, 0.555, 0.437, and 0.210 $\mu\text{W}/\text{cm}^2$ and applied working biases of 0.8, -1.0 , 0.8, and -1.0 V, respectively. The responsivity and detectivity values as functions of wavelength are shown in Figures 2B and S17. CsPbCl₃ responds to light with wavelengths below 410 nm, while the CsPbClBr₂, CsPbBr₃, and CsPbBrI₂ layers respond to wavelengths within 410 and 490, 490 and 520, and 520 and 650 nm, respectively. The linear dynamic ranges of all four layers are over 80 dB (Figure S18), and the noise equivalent powers of the device are calculated to be 3.7×10^{-13} W Hz^{-1/2} at 410 nm, 5.6×10^{-13} W Hz^{-1/2} at 450 nm, 7.0×10^{-13} W Hz^{-1/2} at 520 nm, and 8.6×10^{-13} W Hz^{-1/2} at 635 nm.

We assumed that random fluctuations in the applied current dominate the dark current, and we measured it under a working bias (0.8 V for positive bias and -1.0 V for negative bias). The photoresponse rise time and fall time of the sensors range from 0.85 to 16.2 ms (see Figure S19), and the response times for all photoactive layers increased as the layer thickness

increased. To assess the performance of our device in comparison to the relevant performance of perovskite photodetectors reported in previous works, we present a benchmark in Table S1. The current density-to-voltage curves are displayed in Figure 2C, which reveal that the UV and G diodes operate with a positive bias, while the B and R diodes function with a negative bias, respectively. Given that the open-circuit voltage (V_{oc}) increases with light intensity as a logarithmic function, we opted for relatively higher light intensities for function demonstration and selected 0.8 and -1.0 V as the working biases to obtain stable photoresponse currents and minimal noise from other photoactive layers.

To further understand the color selectivity of the four wavelength ranges, we conducted more color discrimination tests, as shown in Figures 3A–3D. The mixed light sources of UV + B and G + R activate the device separately at the given bias. For instance, Figure 3A shows the photoresponse cycles under UV detection mode with the UV laser being switched on and off while the blue laser stays on. The enlarged pictures in Figure 3A present a negligible difference between dark current density and the current density under high blue irradiance under a working bias of 0.8 V. This implies that blue light barely interferes with the device performance in the UV detection mode. Similarly, the other three detection modes are also demonstrated. Although the current generated by the non-working layer (e.g., the blue layer would be the non-working layer when the UV layer is working) flows against the main current, the current density of this interference is low enough and can be disregarded.

Testing under harsh environmental conditions

Owing to the all-inorganic components of the tetrachromatic image sensor, the device can be used for imaging in harsh conditions, such as under thermal shock and thermal cycling. Temperature-dependent XRD and PL measurements are used to understand the evolution of material structure and optoelectronic properties. Using XRD, we explored the perovskites' lattice distortions and phase transitions under temperature variations ranging from 170 to 570 K (Figure S20). As shown in the Figures 4A–4D, peak shifts for the crystal planes 001, 011, and 002 of the cubic phase at high temperature (or 002, 022, and 004 of the orthorhombic phase at low temperature) can be observed throughout the temperature range, while some other peaks appear and disappear due to the phase transitions.^{22–24} Interplanar spacings of all four perovskites expand as the temperature increases, as shown in Figure S21, which can be seen from the 100 peaks at 15.93° , 15.57° , 15.30° , and 14.80° at 170 K shifting to smaller values at 570 K for CsPbCl₃, CsPbClBr₂, CsPbBr₃, and CsPbBrI₂, respectively. Peaks corresponding to the 111 plane emerge when the materials are cooled to their phase transition points (Figure S22). For example, one peak of CsPbBrI₂ at 41.6° weakens at 415 K and another appears at 44.5° at 240 K. Due to the small temperature range of the tetragonal phase, particularly for CsPbCl₃, we provide a more detailed comparison of the lattice constants and volumes derived from the temperature-dependent XRD in Figure S23, which shows the phase transitions. While positive volume expansion coefficients for crystal units are observed in all mate-

rials, the anisotropic thermal expansion of individual lattice spacings is consistent with existing literature.^{24,25}

The optoelectronic properties of the four inorganic perovskite materials were measured using PL from 140 to 500 K, as shown in Figures 4E–4H. The subtle peak shifts to shorter wavelengths during heating suggest a slight enlargement of the band gap due to the synergistic effects of thermal expansion and electron-acoustic phonon interaction.²⁶ The PL FWHM broadening, as depicted in Figure S24, has been used to estimate the phonon scattering and electron-phonon coupling in semiconductors.^{27,28} Previous studies have suggested that the predominant factor of PL broadening above 100 K is not from acoustic phonon scattering but from electron longitudinal optical phonon interactions.²⁹ This may result in higher electron scattering in perovskite thin films within the high-temperature regime. The peak intensity increase during the cooling process suggests higher radiative recombination and lower defect density, even in the presence of a transition from a high-symmetry crystal structure to a low-symmetry one.³⁰

Responsivity measurements were carried out under various temperatures and pressures (shown in Figures S25 and S26). Pressure-dependent performance showed negligible changes from vacuum to atmospheric pressure. We found distinct steps in the curves indicating the variation of responsivity with temperature in Figures S25A and S25C, while there were no such steps in Figures S25B and S25D. We conjecture that the phase transition process in pure halide perovskite is significantly faster compared to that in mixed-halide thin films. Previous works on solar cells have indicated that the short-circuit density would decrease in the high-temperature regime^{31–33} or stay within a certain range in the low-temperature regime.^{30,34} The divergence in our results may be caused by different test conditions, device components, or structures. We hypothesize that at higher temperatures, injected holes possess greater thermal energy. This increased thermal energy facilitates the holes to drift across potential barriers and allows them to move more easily from the ITO/NiO layer to the perovskite layer at the forward mode junction, resulting in an increase in photocurrent. In addition, the dark state current density exponentially increases with the increased temperature, resulting in the decrease of V_{oc} and the on/off ratio.

To evaluate the operational and thermal stability of the device, we implemented two thermal cycling setups designed to simulate thermal shocks experienced in space applications (refer to Figures S27C and S27D). After running 1,000 cycles by gradually heating to 500 K and cooling to 170 K at a rate of 50 K/min, all four photoactive layers retained a responsivity of >95%. The device experienced less decay during these gradual thermal cycles than when kept at a constant temperature condition (Figure S27A). This is because of the crystalline reconstruction and defect repair after annealing at high temperatures. Even when the device is subjected to a more sudden thermal cycle of heating and cooling by transferring between a hotplate at 500 K and a vacuum environment at 170 K, the device retained >96% of its performance over the test that lasted 20 cycles. We also conducted photoresponse measurements at the extreme low temperature of 16 K (Figure S28), and the device maintained stable operation (responsivity over 95%) throughout the 1 h

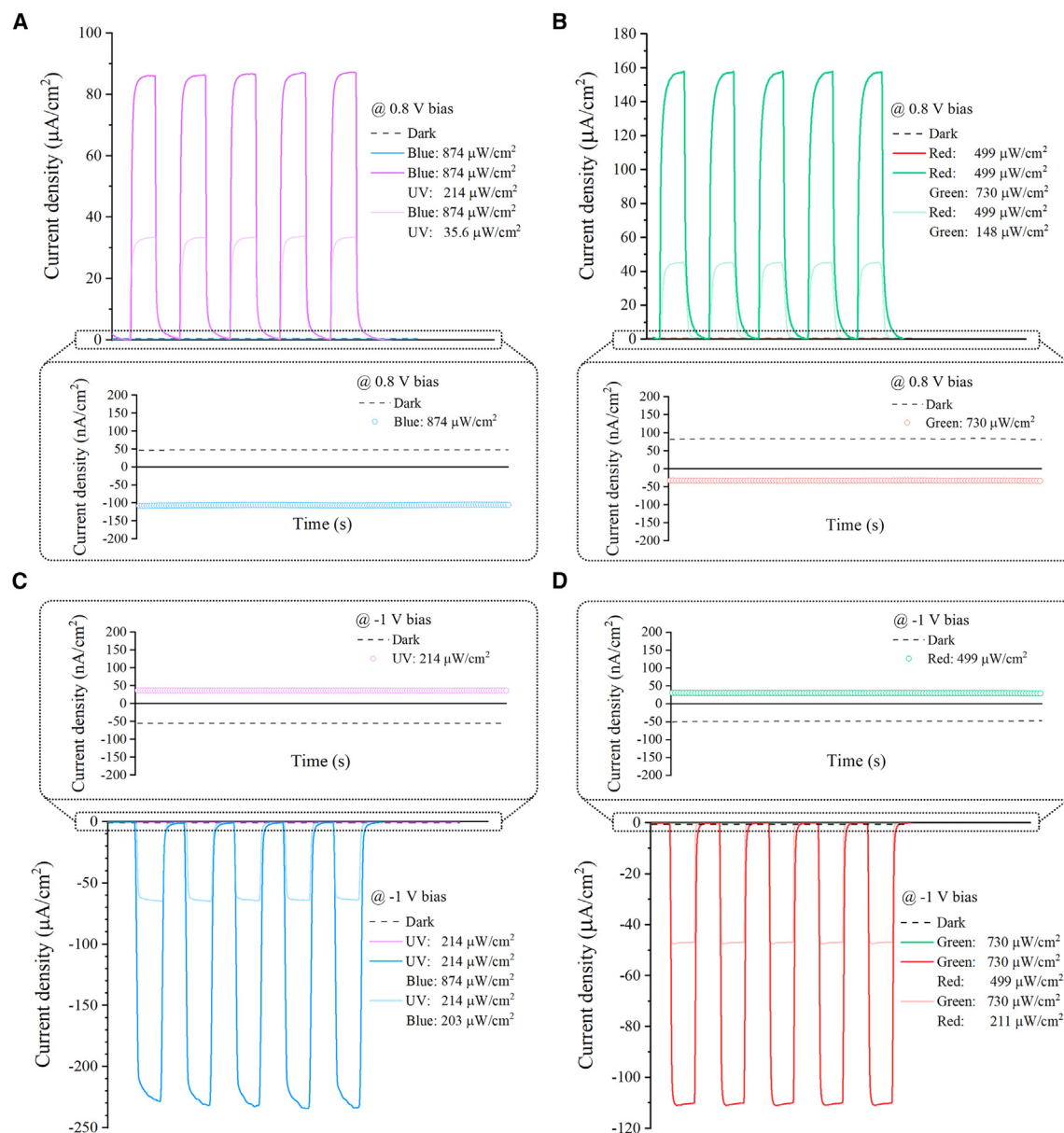


Figure 3. Photocurrent response of mixed light source

Photocurrent response of mixed light sources of 405 and 450 nm (A and C) and 520 and 635 nm (B and D) at forward working mode (A and B) and reverse working mode (C and D), respectively. The enlarged pictures depict the device's current densities under dark and light conditions, showcasing the device's ability to generate low interference signals even in mixed light environments.

measurement. The operational stability in high-temperature regimes (500 K) with the device soaking under various wavelength lasers for over 1 day (shown in Figure S29) reveals only slight performance degradation when operating at high temperatures.

The durability of the device under ambient and vacuum conditions is shown in Figures S30–S32. The estimated T_{90} values, which represent the lifetime at which the responsivity is diminished by 10%, were approximately 1,800, 650, 510, and 380 h, respectively, under vacuum testing for the four layers. While CsPbBr₂ demonstrates good stability under vacuum conditions, it experiences a significant degradation when stored in ambient

conditions. This degradation is primarily attributed to the presence of oxygen and water vapor in the air, which can react with the material and lead to hydrolysis and the subsequent breakdown of the crystal lattice. Furthermore, the specific composition and structure of CsPbBr₂, characterized by a low tolerance factor, render it less chemically stable compared to other halogen perovskite materials.³⁵

To further test the stability of the four perovskite materials under harsh conditions, we heated them up to 200°C and annealed for 100 h. The SEM images of thin films' morphology show negligible differences before and after annealing (Figure S33) except

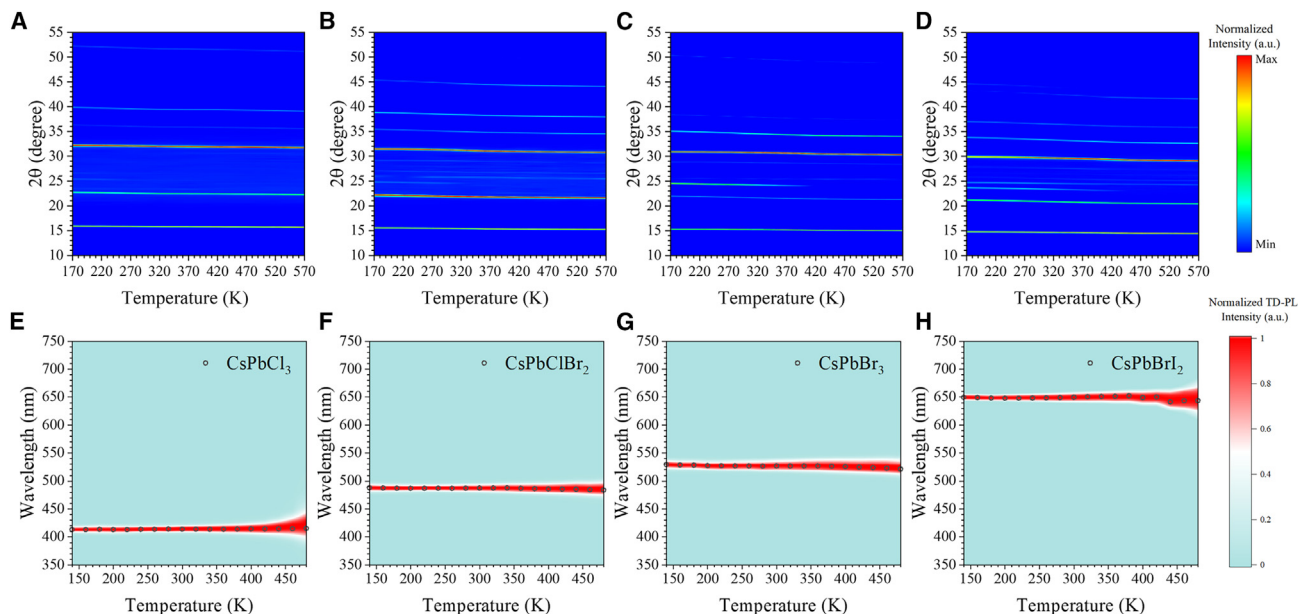


Figure 4. Temperature-dependent XRD (TD-XRD) and PL characterizations of the perovskite thin films

(A–D) TD-XRD contour map of CsPbCl₃, CsPbClBr₂, CsPbBr₃, and CsPbBrI₂ thin films.

(E–H) Contour map of normalized TD-PL spectra of four materials as a function of temperature. The black scatter circles point out the peak positions of each temperature.

for CsPbBrI₂, which is mentioned in the figure caption. As demonstrated in Figures S9 and S10, the XRD peaks of the three CsPbX₃ materials with varying Cl and Br ratios show minimal changes before and after annealing, with no notable formation of additional peaks. In contrast, as indicated in Figures S10C and S10D, CsPbBrI₂ exhibits significant changes in its XRD pattern marked by the appearance of the peaks for CsBr and PbI₂. This reflects a substantial alteration in the crystal structure attributed to the poor phase stability of CsPbBrI₂-type materials, which tend to undergo phase separation and transformation.^{25,36,37} To further examine this, we extracted the halogen vacancy content from the four perovskite materials after the temperature treatment. The results shown in Figure S34 indicate minimal impact on the two Cl-containing perovskite materials, CsPbCl₃ and CsPbClBr₂, with halogen vacancy levels below 2%. In contrast, CsPbBr₃ shows a modest increase in vacancy content (3.52%) after annealing, while CsPbBrI₂ exhibits the largest change with a halogen vacancy proportion of 6.19%, closely correlated with the formation of the additional peaks of decomposition product, proving that the material undergoes significant phase segregation during the annealing process.³⁸ It is crucial to emphasize that halogen vacancies do not imply the loss of halide ions but rather their migration and phase transition behavior.

Tetrachromatic color discrimination and imaging demonstration

To benchmark the device's discrimination capabilities and eliminate chromatic aberration from light sources, we measured absolute light intensities and relative intensity ratios using 405, 450, 520, and 635 nm single-wavelength lasers as light sources. To

control the combinations, we modulated the light intensities. In Figure 5A, each pair of black and white scatter points in the 3D tetrahedral and ternary images are in close proximity, indicating a high degree of resemblance between the reconstructed light combinations and the original light ratios. We further evaluated the variation between actual and detected values in Figure 5B. The blue box (UV/B/G) shown in Figure 5B illustrates a much smaller distance than the other three boxes, which means that the R layer plays a substantial role in the overall deviation (shown as the light pink box UV/B/G/R). Similarly, we inferred that the deviation of each layer occurs in the descending order of R, G, B, and UV layer. This phenomenon is likely caused by the light intensity calibration, as well as scattering and reflection when light passes through the multilayer interfaces. More details can be found in Note S2.

To demonstrate the imaging capabilities of the tetrachromatic image sensor, we created a 4 × 32 × 32 pixel sensor array with a pixel line width of 100 μm and a pixel pitch of 400 μm. The sensor array is integrated with readout circuits and on-chip temperature and pressure sensors, as shown in Figures 6A–6C. We tested the device in vacuum (Figure 6D), low-temperature (Figure 6E), and high-temperature (Figure 6F) environments using images of Earth, Neptune, and Venus from the National Aeronautics and Space Administration (NASA) to simulate the space environment, and an image of the Crab Nebula from NASA was used as the target in the ambient environment. We also used the sensor array to image a fuel stove, which can emit UV radiation (Figure 6G) at high temperature. We also used a filtered UV light-emitting diode backlight and patterns to demonstrate the UV channel's functionality further. The UV, B, G, and R channels are displayed separately to assess the image quality of each

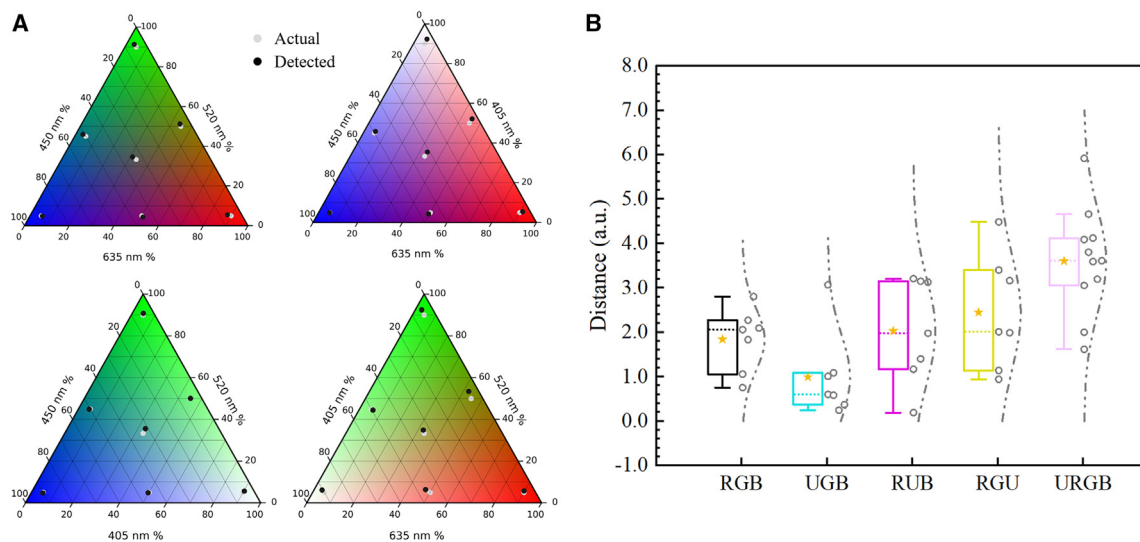


Figure 5. Capability of the tetrachromatic image sensor system to discriminate mixed lights

(A) Ternary light combination maps based on 405, 450, 520, and 635 nm light sources.

(B) Standard deviation of five maps that show the deviation of actual light combination and detected light combination after calculation.

channel (Figures S35 and S36). All channel figures were corrected according to the temperature coefficients from the temperature-dependent data of responsivity (Figure S25) in the image output process. The original images of low- and high-temperature demonstrations are presented in Figure S36. Small deviations were observed between the real and reconstructed images in a vacuum, and the deviations are more noticeable at high temperatures, corresponding closely to the temperature coefficients. Considering this, we installed on-chip sensors to provide feedback for calibrating the output data to produce better results (see Figure 6H). The UV/B channels exhibit higher accuracy than the R/G channels, which is consistent with the light discrimination results as aforementioned.

Conclusion and outlook

We have developed an all-in-one tetrachromatic image sensor system with a $4 \times 32 \times 32$ pixel configuration. The sensor uses two electrically switchable color-selective p-i-n-i-p structures to discriminate four wavelength ranges by using four perovskite layers with tunable band gaps. We measured the effects of temperature and pressure on the phase transition, lattice distortion, and optoelectronic performance of the four perovskites. Our research aims to enhance the performance and stability of various perovskite sensors for working in harsh environmental conditions. The tetrachromatic sensor can reproduce high-accuracy images under harsh environments even after being subjected to thermal shock cycles and changing pressures. However, the challenges of crosstalk and pixel size persist as the primary obstacles to miniaturize the device and increase its resolution. To address these challenges, we are investigating approaches including the incorporation of insulating layers,^{39,40} electrode isolation,¹¹ and optimization of electrode geometry. Additionally, we are exploring the potential of using thin-film transistors and complementary metal-oxide-semiconductor substrates as viable

alternatives to crossbar electrodes. These advancements provide a possibility of fabricating high-resolution pixel arrays in the near future. This device may find applications in space exploration, where sensors are required to be remain stable and durable under extreme temperatures and vacuum conditions while providing the ability to detect a wide range of wavelengths, including visible light, UV light, and other parts of the electromagnetic spectrum. This device offers a wide spectral response and high sensitivity, enabling the detection and analysis of diverse electromagnetic radiation wavelengths, and may also find uses in self-driving cars, artificial retinas, and industrial system monitoring.

EXPERIMENTAL PROCEDURES

Resource availability

Lead contact

Requests for further information, resources, and reagents should be directed to and will be fulfilled by the lead contact, Zhiyong Fan (eezfan@ust.hk).

Materials availability

This study did not generate new unique reagents.

Data and code availability

The data that support the findings of this study are included in the main text and the supplemental information. More data are available from the corresponding author upon reasonable request. The source codes for MATLAB and Python are available from the corresponding author upon reasonable request.

Materials

All chemicals were purchased from Xi'an Polymer Light Technology and used as received without further purification.

Simulations

Below are the details of the optical simulation conducted for the design of the tetrachromatic all-inorganic perovskite image sensor. For the optical simulation for the design of the tetrachromatic image sensor, a numeric computing environment developed by MathWorks (Zenodo link: <https://doi.org/10.5281/zenodo.10781549>) (MATLAB) was used to simulate the transmission

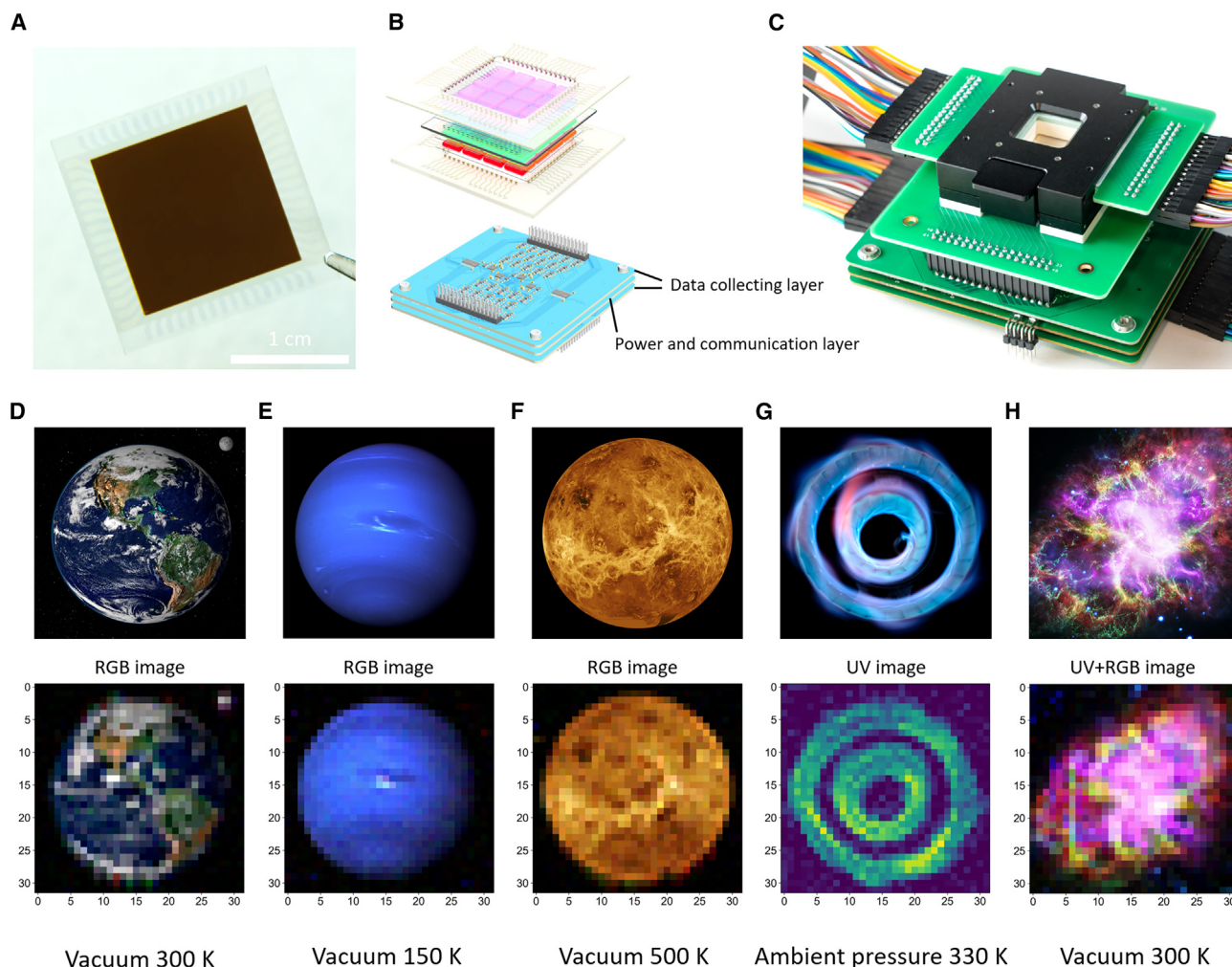


Figure 6. Image detection demonstration in the extreme environment

(A) Image of the four-layer device.

(B and C) Schematic (B) and image (C) of the tetrachromatic image sensor system.

(D–H) Photographs and captured images of Earth (D), Neptune (E), Venus (F), flame (G), and Crab Nebula (H) in different environments (labeled in the images). Notably, the fuel stove emblazened the flame, and the projector projected the other pictures. The calculated output image of flame detection was UV channel, while other images were RGB channel.

versus the wavelength and thickness of different perovskite materials (see [Figure S2](#)) based on the refractive index and absorption (n , k) measured by the ellipsometer (J.A. Woollam M-2000VI). The simulation is based on the multi-layer interference theory, which is widely used in designing antireflection coating. By adding the absorption term of the refractive index and the refraction factor of the incident angle, the reflective and transmissive spectra are influenced by the real and imaginary parts of the index of refraction layer by layer. Then, the angular dependence of the image sensor, especially the cross-talk between the neighboring pixels, is calculated (see [Figure S3](#)). Based on those simulation results, the performance of the tetrachromatic image sensor is designed with a critical thickness for different layers including all the layers in the sensor.

Fabrication of tetrachromatic all-inorganic perovskite image sensor

Double-sided ITO-patterned (ITO thickness is 120 nm on each side) quartz plates (100 μm line width and 400 μm pitch as shown in [Figure S12](#)) were cleaned in sequence with detergent, deionized water, ethanol, acetone, and isopropyl alcohol by an ultrasonic washer. Before use, ITO-coated substrates

were dried by N_2 and exposed to O_2 plasma for 10 min. More details about the fabrication processes can be found in [Note S1](#).

Characterization of material and photoresponse

Complex refractive indices (n , k) and UV-vis absorption and transparency spectra of four perovskite materials were obtained using J.A. Woollam M-2000VI Spectroscopic Ellipsometer and Lambda 1050+ (PerkinElmer). Surface morphology of thin films was carried out on the Dimension ICON (Bruker). A field-emission SEM (JSM-7100F, JEOL) was used to characterize the device's top view and cross-section view under an acceleration voltage of 5 or 10 kV. The XPS and UPS spectra were recorded on a PHI 5000 Versa Probe III. TD-XRD spectra were derived from an X-ray diffractometer (Rigaku SmartLab, 9 kW, Cu target, BB geometry). Temperature-dependent PL measurements were carried out on the QE-Pro spectrometer (Ocean Insight) and Newton 920 CCD (Oxford Instruments). The emission wavelength used in the measurements was 266 nm, and the sample temperatures were controlled by the Linkam Optical Shearing System (CSS450). As for the photoresponse characterization, we used an optical power meter (Thorlabs, PM100D) to

calibrate the light intensity. The 3 and 30 mW lasers of 405, 450, 520, and 635 nm controlled by a pulse-width modulation controller were used as single-wavelength light sources, and we used neutral-density filters to adjust the light intensities. A xenon lamp with grating served as a wavelength-tunable light source (300–900 nm). The current-time curves and current-voltage curves of devices under high- and low-temperature conditions were measured by TTPX Cryogenic Probe Station, Lake Shore model 336 Cryogenic Temperature Controller, and a Keithley 2450 source meter and with a liquid nitrogen tank. All temperature-dependent measurements had a temperature step and holding time of 20 K and 10 min, respectively.

Integration of tetrachromatic all-inorganic perovskite image sensor system

We designed the test system as shown in Figures S36 and S37. For both UV/B and R/G layers, we used two 16-channel multiplexers (ADG1606) for row selection. In each column, there was one transimpedance amplifier (based on AD8615), one amplifier (AD8615), and one 16 bit analog-to-digital converter (ADC; ADS1110). All 32 ADCs were connected in series with an I2C bus for output. The microcontroller unit (MCU) used in this design was the ESP WROOM 32D. This MCU was responsible for scanning the photodetector arrays, processing the signals, and communicating the data to external devices. The I2C bus connected the MCU to the ADCs, enabling seamless data transfer. The printed circuit board was also embedded with one temperature sensor (DS18B20U) and one pressure sensor (BMP280) to monitor environmental conditions (Figure S39).

Demonstration under normal and harsh conditions

We calibrated every pixel by taking dark and flat files under room temperature (298 K), and flat-field images were taken under a 3 mW/cm² light source. The images were projected on the image sensor by a projector with two convex lens sets. The image detected under normal conditions (room temperature, atmospheric pressure) is reconstructed by the Python program based on readout signals. Furthermore, a microheater was used to heat the image sensor to 200°C to simulate the harsh heat environment, while a vacuum chamber with circulating liquid nitrogen cooled the device down to –100°C to simulate harsh cold circulation. All aforementioned harsh condition measurements were carried out under 5 × 10^{–2} Pa.

SUPPLEMENTAL INFORMATION

Supplemental information can be found online at <https://doi.org/10.1016/j.device.2024.100357>.

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AUTHOR CONTRIBUTIONS

Z.F. and X.Q. conceived the ideas and designed the experiments of this work. X.Q. and Y.D. fabricated the device, integrated the demo system, and reconstructed the images. H.J. and X.Q. carried out the XRD data analysis and refinement. Z.S. and X.Q. contributed to the optical characterizations and sim-

ulations. X.Q., H.J., Y.Z., P.W., B.R., K.Z., Z.L., Y.B.C., B.L., J.W., and Y.L. performed the materials characterizations for perovskite and metal oxide thin films. X.Q., Z.F., Y.Z., S.P., Y.D., Z.L., B.R., Y.B.C., and Z.M. wrote and revised the manuscript. All the authors discussed the results and commented on the manuscript.

DECLARATION OF INTERESTS

We, the authors, have a pending patent related to this work: “Tetrachromatic Perovskite Image Sensor,” US patent, application no. 63/587,148, filing date: Oct. 1, 2023.

DECLARATION OF GENERATIVE AI AND AI-ASSISTED TECHNOLOGIES IN THE WRITING PROCESS

During the preparation of this work, the authors used ChatGPT-3.5 Turbo in order to polish the language. After using this tool, the authors reviewed and edited the content as needed and take full responsibility for the content of the publication.

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