

Multimodal X-ray Detection Enabled by Ion–Electronic Dynamics in Hybrid Perovskites

Yalu Li, Can Zou, Da Liu, Zhanpeng Wei, Xie Zhang, Hua Gui Yang, Yu Hou,* and Shuang Yang*

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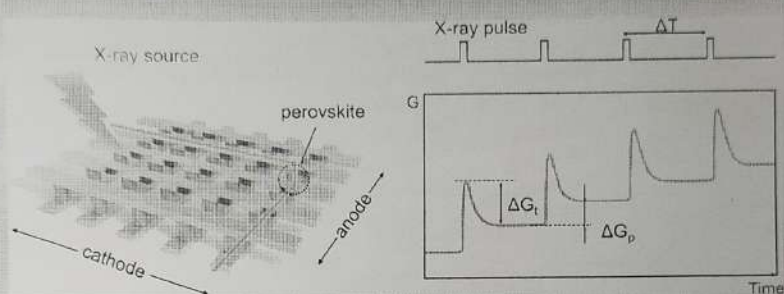
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ABSTRACT: X-ray dosimetry and imaging have been utilized in a wide range of applications. However, the complex components of detection, memory, and computing units of commercial detection systems inevitably make it structurally complex and energy consuming. Here, a multimodal two-terminal perovskite device is demonstrated that can measure real-time dose rate and record the accumulated dose of X-ray pulse signals concurrently by decoupling volatile electronic and non-volatile ionic variables of the conductance (G). The device delivers an X-ray sensitivity of $1159 \mu\text{C Gy}^{-1} \text{cm}^{-2}$, a limit of detection (LoD) of 27.9 nGy s^{-1} , a limit of memory (LoM) of $0.32 \mu\text{Gy s}^{-1}$, as well as multiple non-volatile storage states of around 40 nS . As a proof-of-concept, a perovskite crossbar array integrates the functions of sensitive X-ray detection, multi-state data memorization, as well as image pre-processing, offering a promising platform toward intelligent, robust, and lightweight radiation imaging systems.

The discovery of X-ray photons has revolutionized a wide range of scientific fields, from security inspection and industrial radiography to diagnostic imaging.^{1–5} The existing X-ray imaging system is based on the Von Neumann architecture comprising physically separated X-ray detectors, data memories, and processors, in which the long-distance data conversion and communication would generate a large amount of redundant data together with high energy consumption, delayed computational tasks, and cumbersome hardware.⁶ With the data expansion of machine vision and artificial intelligence techniques, there is a growing requirement to eliminate the sensor, memory, and processor interface in disruptive architectures.⁷

In recent years, neuromorphic sensor devices, like optoelectronic resistive random-access memories (ORRAMs) and synaptic phototransistors (SPTs), have emerged as the next-generation paradigm that can combine the sensing, memory, and processing functions of optical stimulus at a single-unit-device scale.^{8,9} Under X-ray irradiation, active materials, like SrTiO_3 , have shown a persistent photoconductivity (PPC) effect, which opens possibilities to tackle these limitations.¹⁰ However, the integration of these functionalities has been generally made a trade-off in device performance. For example, the response current of synaptic photodetectors shows resemblance to short-term plasticity (STP) and long-term potentiation

(LTP) behaviors of human synapses, leading to the inability to accurately quantify the radiation intensity.¹¹ In addition, the ionic–electronic signals in these devices often interweave with each other, whose sophisticated dynamics impose a great challenge to resolve the dynamic radiation signals.¹²

Here, we explore the X-ray-induced electronic–ionic dynamics of a polycrystalline perovskite device with a vertical electrode/semiconductor/electrode configuration. We observe two conductance channels in the perovskite device, i.e., a transient X-ray-induced electronic current similar to signals in conventional semiconductor detectors and a persistent conductance change caused by radiation-doping behavior. We demonstrate that both the transient and persistent photoconductance channels respond rapidly to X-ray pulses and can be deconvoluted to resolve the dose rate and the accumulated radiation history of X-ray photons. The integrated crossbar array enables real-time imaging and recording of X-ray signals paired with a

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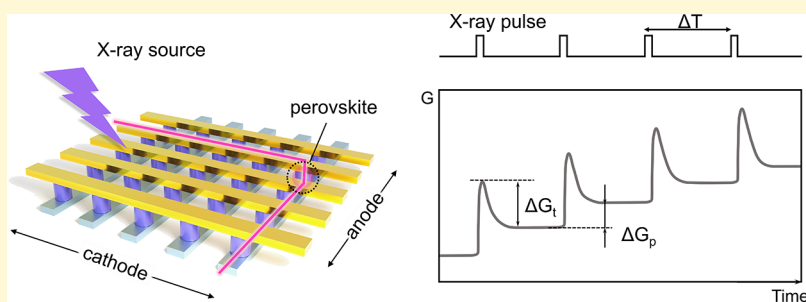
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contrast enhancement function, showcasing its potential all-in-one utilization in sensitive energy-efficient X-ray imaging, storage, and computing applications.

The light–matter interaction between X-ray photons and perovskite materials is shown schematically in Figure 1a. When an X-ray photon interacts with a semiconductor, several fundamental mechanisms are involved, including photoelectric effect, Compton scattering, and electron–positron pair production, in which the first one serves as the primary mode at photon energy levels below 100 keV in our study.^{13–15} This process begins with the ejection events of high-energy electrons, which interact with the semiconductor via inelastic scattering mechanisms and lead to secondary excitations and the generation of

multiple electron–hole pairs.¹⁶ Under an electrical bias, the electron–hole pairs are separated and collected by the respective electrodes, permitting a direct measure of the incident X-ray photon flux by the transient conductance signal (ΔG_t) (Figure 1b).

Another possible interaction mechanism is the ionization or lattice displacement in solids under high-energy radiation. For hybrid perovskites, the structural evolution under X-ray exposure has been identified to be dose-dependent before complete radiolysis: the concentration of structural defects or decomposed phases is often correlated with the accumulating X-ray dose.^{17,18} Instead, the change of defect density may be employed as a variable for evaluating the cumulative dose of

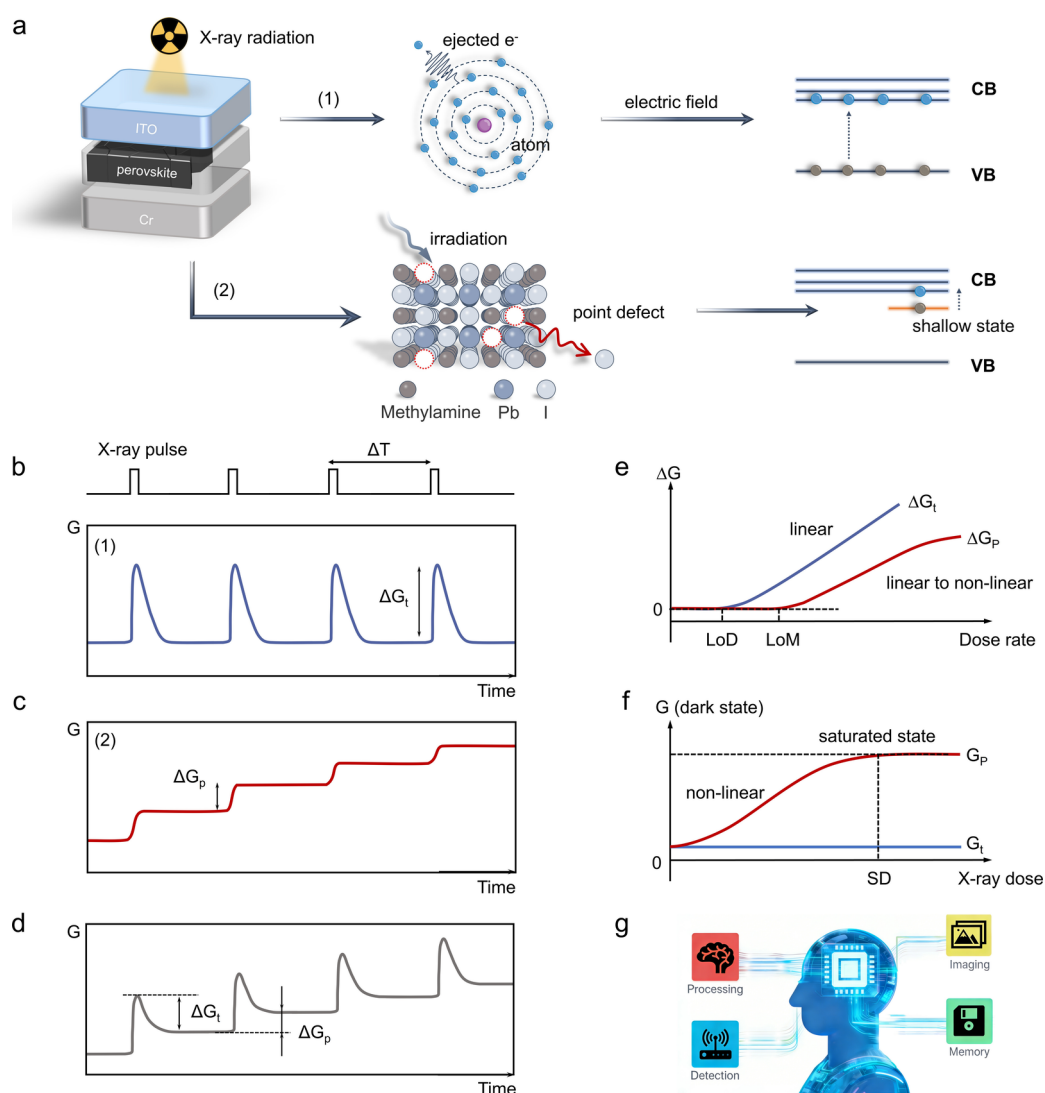


Figure 1. Electronic and ionic response under X-ray irradiation. (a) Schematic illustration of the multimodal two-terminal detector structure and its two X-ray response pathways. Pathway (1) represents the direct X-ray detection of the photoelectric effect. The blue spheres denote photoelectrons. Pathway (2) depicts the X-ray-induced electronic–ionic dynamics. The white circles represent iodine vacancies (V_I). (b) Time-dependent conductance evolution of the detector governed by the photoelectric effect (pathway 1), corresponding to the transient conductance change (ΔG_t). (c) Time-dependent conductance evolution of the detector dominated by X-ray-induced ionic defect dynamics (pathway 2), corresponding to the persistent conductance change (ΔG_p). (d) Overall time-dependent conductance response of the multimodal two-terminal detector under X-ray irradiation, highlighting the coexistence of transient detection and persistent memory functions. (e) Dependence of the conductance change on the X-ray dose rate. The blue and red curves represent ΔG_t and ΔG_p , respectively. LoD and LoM denote the limit of detection and the limit of memory. (f) Conductance as a function of accumulated X-ray dose. The blue and red curves correspond to the transient conductance (G_t) and persistent conductance (G_p), respectively. SD refers to the saturation dose of G_p under X-ray exposure. (g) Multifunctionally integrated two-terminal detector with the detection, imaging, processing, and memory functions.

radiation. One known example is the metal–oxide–semiconductor field-effect transistor (MOSFET) dosimeter, where radiation creates defects in the oxide layer and determines the threshold voltage with respect to the deposited dosage.^{19–21} Given the unique defect chemistry in perovskites, X-ray photon irradiation would modulate the local crystalline structure and thereby result in the persistent conductance change (ΔG_p) under certain dose rates,²² which may enable the dose-dependent memory behavior in perovskite devices (Figure 1c).

When both electronic and ionic contributions are considered, the overall conductance change (ΔG) can be decomposed into ΔG_t and ΔG_p in perovskite devices (Figure 1d,e). The electronic ΔG_t response shows a linear dependence on X-ray photon flux, together with a limit of detection (LoD) at a minimal dose rate, which manifests the typical current mode similar to direct semiconducting detectors. Meanwhile, the non-volatile nature of ionic ΔG_p response behaves as an optoelectronic resistive switching behavior, analogous to the operation of a radiation dosimeter. Since the activation of ionic species is generally more difficult than charge carriers in hybrid perovskites,²³ ΔG_p is expected to appear over a high dose-rate threshold, defined as the limit of memory (LoM), which can filter the weak X-ray signals and realize the pre-processing of the radiation information. Likewise, the total conductance (G) of the device in the dark state consists of transient conductance (G_t) and persistent conductance (G_p): the former returns to the initial value after X-ray irradiation is switched off, whereas the latter increases and eventually saturates with X-ray photon deposition (Figure 1f). The temporal discrepancy of the two channels may permit decoupling of electronic–ionic signals, which creates possibilities to integrate different functionalities into a single device, including detection, data storage, and signal processing (Figure 1g).

We fabricated multimodal perovskite devices with a two-terminal configuration of Cr/MAPbI₃/ITO (Figures S1 and S2). We selected Cr as the top electrode for its chemical stability and polycrystalline MAPbI₃ perovskite as the active layer because of its simple solution fabrication and strong X-ray attenuation with high-Z compositions. In contrast to conventional materials like metal oxides and sulfides with large bond strength, the Pb–I bonds in MAPbI₃ are much weaker and can readily dissociate under high-energy radiation²⁴ and may provide an underlying physical basis for X-ray-modulated memristive behavior (Figures S3 and S4).

Electrical characterizations were first performed in the absence of X-ray irradiation to examine the intrinsic memristive properties of the devices. When a voltage sweep was applied to the ITO electrode while the chromium electrode was grounded, reproducible and reversible bipolar switching between high-resistance and low-resistance states (HRS and LRS) was observed at low operation voltages around 1 V (Figure S5). Similar switchable resistance phenomena have been universally demonstrated in other perovskite materials and with various top electrodes, such as nickel, gallium, and platinum.²⁵ Upon X-ray exposure, a pronounced reduction in the threshold voltage required for resistive switching was observed, indicative of the possibility of X-ray-induced memristive behavior in perovskite devices. The robustness of this radiation-assisted effect was verified by stable operation over 100 consecutive switching cycles and consistent behavior across 20 independent devices (Figures S6 and S7). A more convincing indication of the radiation effect in resistive switching is the inverse correlation

between the set voltage and X-ray dose rate (Figure S8), which substantiates the dominant role of radiation in generating the conductive channel.

To further elucidate the impact of X-ray irradiation on the dynamic electrical response, the device was exposed to X-ray pulses from the ITO side under a constant read voltage of 0.05 V. Figure 2a presents the conductance evolution of the devices under a series of short X-ray pulses ($\Delta T = 2$ s, pulse width = 0.2 s) with moderate dose rates ranging from 0.15 to 2.12 $\mu\text{Gy s}^{-1}$. We also scrutinized the ΔG_p dependence of the perovskite device under low X-ray dose rates (Figures 2b and S9). We observed the appearance of persistent photoconductivity at a threshold dose rate of 0.32 $\mu\text{Gy s}^{-1}$, corresponding to the proposed LoM value. At a low dose rate of 0.15 $\mu\text{Gy s}^{-1}$, only transient conductance changes that rapidly followed the X-ray signal were detectable, indicating pure detection behavior below the LoM regime. As the dose rate increased to 0.79 $\mu\text{Gy s}^{-1}$, the device underwent both photoconductive and memristive behaviors, characterized by stepwise increases in conductance with each X-ray pulse. Enlarged views of the X-ray response reveal that conductance gain involves two components: the transient ΔG_t falls back immediately after the X-ray is off, whereas the persistent ΔG_p is retained until the next pulse, consistent with the proposed electronic–ionic conduction mechanism. Continuous X-ray pulses resulted in the gradual saturation of permanent conductance at approximately 40 nS due to the thermodynamic limitation, beyond which further irradiation only produced transient conductance changes. A further increase in dose rate up to 2.12 $\mu\text{Gy s}^{-1}$ manifested similar mixed electronic–ionic dynamics in electrical conductance, which culminated in an accelerated overall saturation effect of ΔG_p (Figure S10). Figure S11 verifies that this decoupling phenomenon is independent of the specific pulse shape.

Figure 2a sketches the evolution of the total peak conductance value G as a function of irradiation time of the perovskite device, where the electronic and ionic components cannot be quantified straightforwardly. We therefore deduced the persistent components, i.e., the stepped baseline, to isolate the transient electrical response and summarized the ΔG_t values of the perovskite device in Figure 2c. Notably, accurate signal extraction in this process is predicated on the assumption of a stable baseline without drift. It can be seen that ΔG_t is linearly correlated with the dose rate and independent of the non-volatile memory behaviors, remaining stable across the entire test, which shows resemblance to the typical direct X-ray detectors. In view of the transient current variation under radiation, the sensitivity and detection limit were quantitatively determined to be 1159 $\mu\text{C Gy}^{-1} \text{ cm}^{-2}$ and 27.9 nGy s^{-1} , respectively, under a very low electric field of 0.05 V ($0.1 \text{ V } \mu\text{m}^{-1}$) (Figure 2d,e). The photocurrent responses at low X-ray dose rates are shown in Figure S12. Such competitive detection performance is a vital prerequisite for stable, low-power ion–electronic dynamics without field-induced degradation.²⁶

Unlike the transient conductance change that shows linearity to X-ray photon flux, the persistent ΔG_p showcases a synaptic depression behavior: the variation of persistent conductance under the identical X-ray pulse obviously declined under incremental accumulated dose (Figure S13). As a consequence, the conductance was gradually saturated at an X-ray dose of $\sim 10 \mu\text{Gy}$ (Figure 2f). Particular emphasis is placed on the multiple stable conductance states during these experiments, as interpreted in Figure 2g, where the perovskite device operated

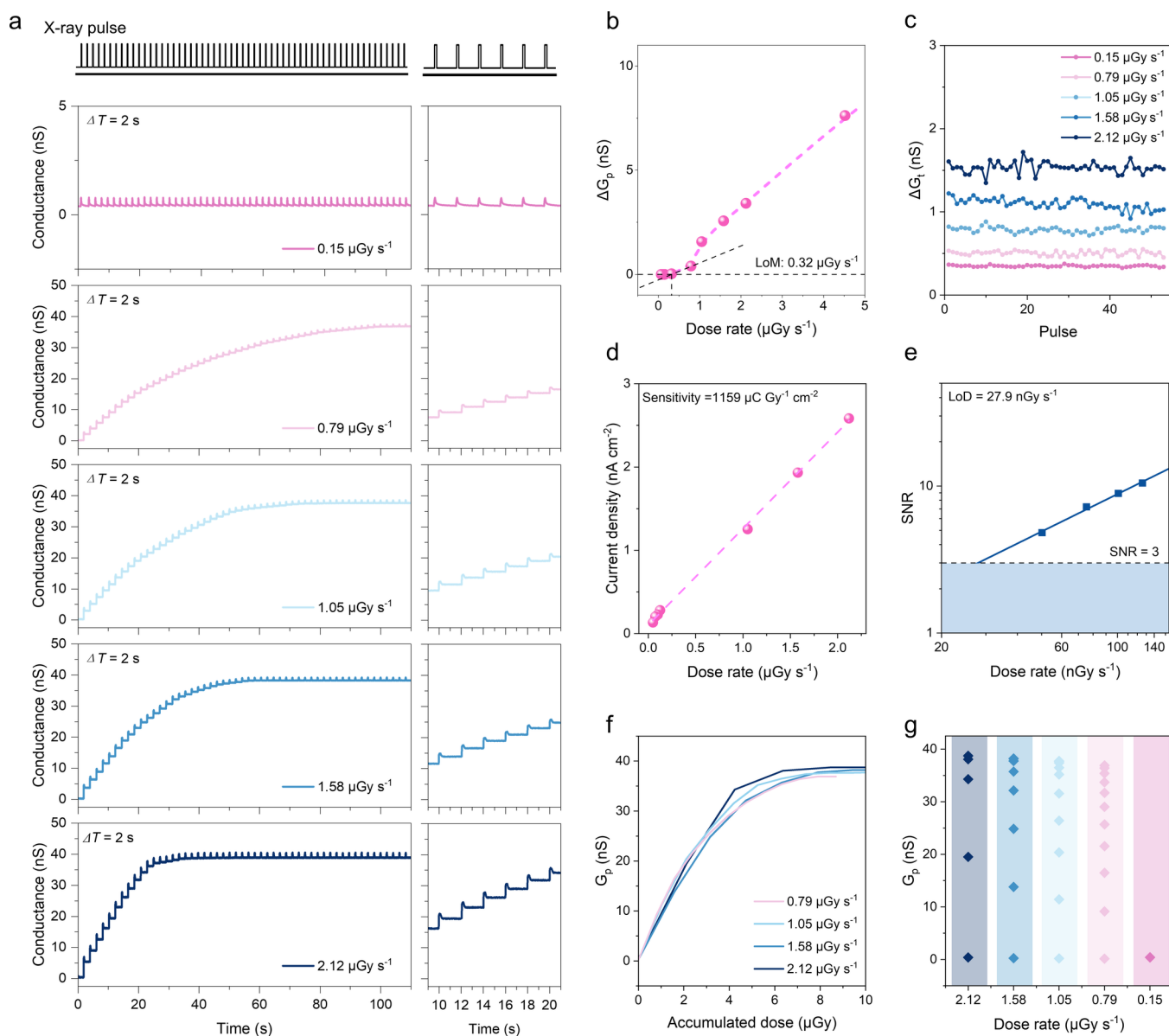


Figure 2. Characterizations of X-ray-induced multimodal perovskite devices. (a) Time-dependent conductance curves of detectors under radiation pulses with different dose rates ($0.15\text{--}2.12 \mu\text{Gy s}^{-1}$). The right panel magnifies the six pulse responses recorded within the same time window under different dose rates. (b) Dependence of ΔG_p as a function of dose rate. LoM is $0.32 \mu\text{Gy s}^{-1}$. (c) Relationship between ΔG and pulse number under different dose rates. (d) Current density as a function of the incident dose rate for the detector under a 0.05 V bias. (e) Dose-rate-dependent signal-to-noise ratio (SNR) of the device measured under a 0.05 V bias. LoD is obtained from the fitting line at an SNR of 3. (f) G_p as a function of accumulated dose. No G_p is generated under an X-ray dose rate of $0.15 \mu\text{Gy s}^{-1}$, suggestive of the potential of signal pre-processing of image data. The storage of irradiation shows a saturated X-ray dose of $\sim 10 \mu\text{Gy}$. (g) Relationship between the number of pulses and the dose rate to reach the saturated conductance state. Each data point corresponds to five pulses.

under 0.79 , 1.05 , 1.58 , and $2.12 \mu\text{Gy s}^{-1}$ encompassed 52, 34, 28, and 15 storage states, respectively. The observed dose-rate-dependent, stepwise conductivity modulation under X-ray pulses highlights the potential of perovskite devices for multi-level data storage and radiation-programmable memory applications.

To shed light on the memristive mechanism of the perovskite device, we operated conductive atomic force microscopy (c-AFM) measurements to spatially resolve the conductivity variations in perovskite thin films subjected to different X-ray doses. The surface topology and local dark current were

simultaneously mapped by a cantilever equipped with a current-to-voltage preamplifier. As shown in Figure 3a,b, the perovskite films exhibit relatively homogeneous conductance within each grain, regardless of the X-ray doses, which excludes the possibility of conductive filament formation when using active metal electrodes.²⁷ Upon the accumulation of incident X-ray photons, there was an overall enhancement in conductance within the perovskite grains; however, the current at the boundary regime remained unchanged. Consistent with this observation, the perovskite films retained the polycrystalline morphology after irradiation, as shown in Figure S14.

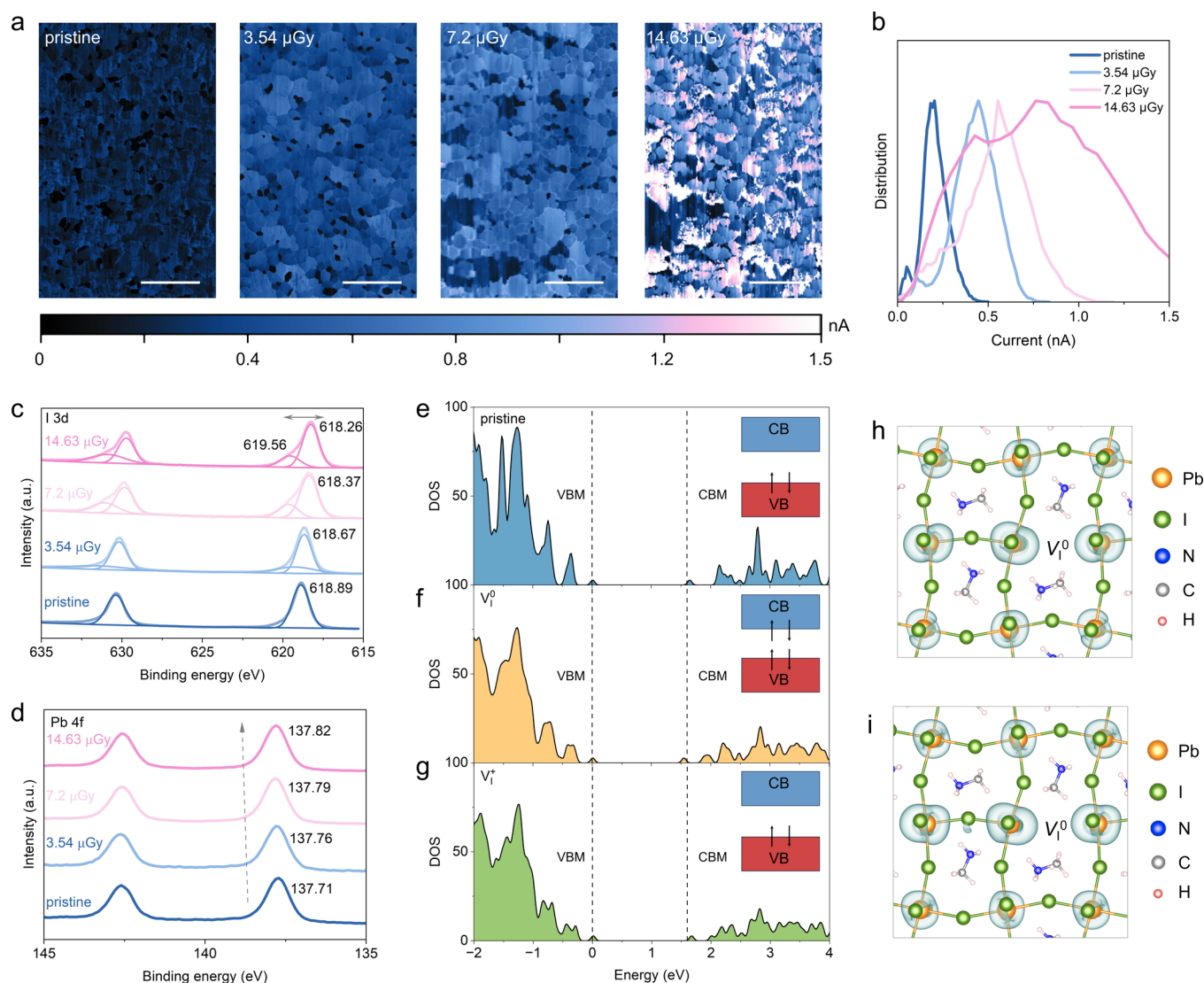


Figure 3. Mechanism of the memristive perovskite devices. (a) Conducting-atomic force microscopy (c-AFM) images. Scale bars are 1 μm . (b) Corresponding current distribution of the perovskite film after exposure to different X-ray doses. XPS spectra of (c) I 3d and (d) Pb 4f after exposure to different X-ray doses. (e–i) Formation of shallow trap states in the density of states.

Therefore, the memristive behavior of conductance should stem from the bulk perovskite grain, other than grain boundaries, surfaces, or extrinsic impurities.

In metal oxide materials, non-volatile conductivity changes driven by light irradiation are often attributed to defect generation, such as oxygen vacancies, which impart shallow electronic states near band edges.^{28–30} We therefore employed X-ray photoelectron spectroscopy (XPS) to probe the associated chemical structure and valence state of perovskite films under X-ray doses in the range of 0–14.63 μGy . The metal electrodes were delaminated to expose the perovskite surface for the measurements. Upon X-ray irradiation, the main I 3d_{5/2} peaks gradually shift from 618.89 to 618.26 eV, accompanied by the emergence of another component at $\sim$ 619.56 eV (Figure 3c,d), which resembles the XPS results of vacancy-type defects in metal oxides and sulfides.³¹ The Pb 4f spectra comprise two contributions, for example, 4f_{5/2} and 4f_{7/2}, located at 142.54–142.61 and 137.71–137.82 eV, respectively. Steady-state photoluminescence (PL) and time-resolved PL (TRPL) measurements

revealed the negative correlation between PL intensity and X-ray dose, indicating the defect formation in perovskites under X-ray radiation (Figures S15 and S16).

It has been known that halide vacancies are ubiquitous in the perovskite crystal lattice, as intensively validated by experimental characterizations, like integrated differential phase contrast (iDPC) scanning transmission electron microscopy (STEM).³² Previous computational studies have signified the low formation energy (0.29–0.87 eV) and the shallow defect nature of iodine vacancies in perovskites.^{33–36} We therefore carried out first-principles calculations based on the density functional theory to encode the electronic structure of MAPbI₃ without and with V_I under different charge states (Figure 3e–i). In Figure 3e–g, we show the density of states for MAPbI₃ containing V_I^0 and V_I^+ in comparison with that of pristine MAPbI₃. The presence of V_I^0 introduces a Kohn–Sham level that is closely resonant with the conduction-band minimum (CBM), which is because the CBM is primarily comprised of iodine p orbitals. The iodine vacancies induced electronic states that are derived from the

CBM in a perturbative manner. This Kohn–Sham level becomes unoccupied from transitioning from V_1^0 to V_1^+ (Figure 3g). We visualize the wavefunctions of the Kohn–Sham level (two degenerate bands) associated with V_1^0 in Figure 3h,i. Apparently, these

wavefunctions are highly delocalized, meaning that the electrons carried by V_1^0 can easily be ionized and donate to the lattice. These features imply that V_1 is a shallow donor, which helps to improve the n-type conductivity of the system.³⁷

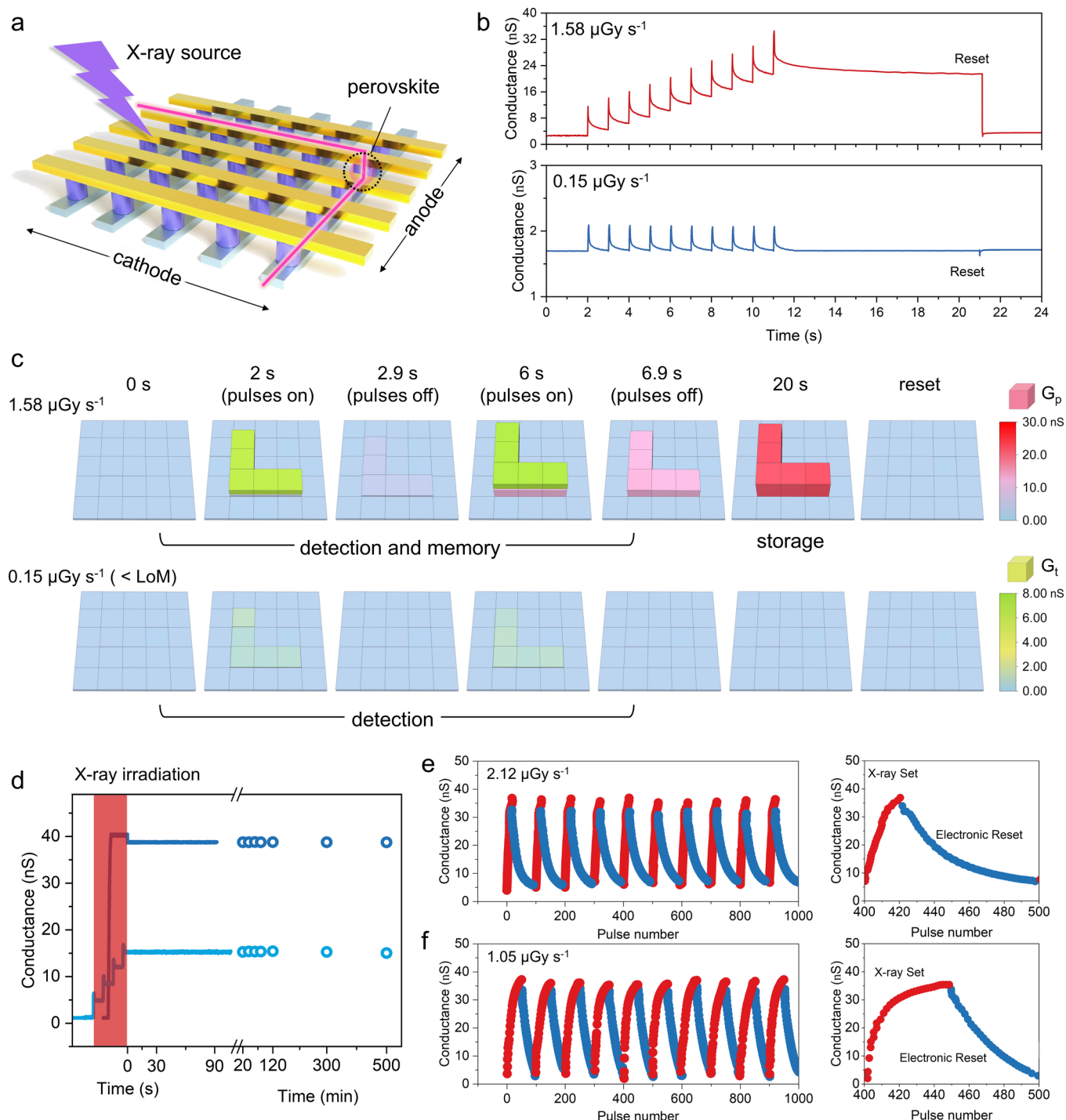


Figure 4. X-ray imaging and memorizing of the memristive perovskite devices. (a) Schematic illustration of the 5×5 crossbar array structure. (b) SET and RESET processes of a pixel measured under dose rates of 1.58 and $0.15 \mu\text{Gy s}^{-1}$. (c) Image detection and memory read at different times under a 0.05 V read bias. The top row corresponds to a dose rate of $1.58 \mu\text{Gy s}^{-1}$, and the bottom row corresponds to a dose rate of $0.15 \mu\text{Gy s}^{-1}$ ($< \text{LoM}$). The detection signal consists of G_t and G_p . This dose rate-dependent response preprocesses the X-ray images by filtering the weak information below LoM. (d) Non-volatile conduction changes of the multimodal device under different X-ray inputs. Multi-level reversibility of the multimodal device under the dose rate of (e) $2.12 \mu\text{Gy s}^{-1}$ and (f) $1.05 \mu\text{Gy s}^{-1}$. The red curves represent the SET process, and the blue curves represent the RESET process.

On the basis of the above results, we conclude that the memristive channel in the multimodal perovskite device operates via a radiation-induced doping mechanism. In this process, X-ray irradiation promotes the formation of V_i defects, which introduce shallow states near band edges and, thereby, proliferate the carrier density for improved persistent conductivity. Unlike conventional semiconductors, the unique defect tolerance of perovskites to the vacancy-type point defects allows the retention of the semiconductive property³⁸ and will not notably affect the transient operation channel for direct X-ray detection through the transient current signal.

Having manifested the integrated functionalities of multimodal perovskite devices that are functionally analogous to those of the human retina, we constructed a 5×5 crossbar array (Figure 4a) to demonstrate how this device senses, preprocesses, and memorizes X-ray information. In this architecture, each cell corresponds to an image pixel. X-ray image perception and memorization experiments were operated at dose rates of 1.58 and 0.15 $\mu\text{Gy s}^{-1}$, as illustrated in Figure 4b. An “L”-shaped mask was used to define the irradiance of X-ray photons. The stimulus protocol consisted of three sequential steps: (1) ten X-ray pulses with a period $\Delta T = 1$ s, starting at 2 s; (2) a dark interval of 10 s after X-ray irradiation; and (3) electrical reset by applying a reverse bias of -0.5 V.³³ Before applying the stimulation, all devices were initialized at HRS. Current signals from all pixels were recorded in real time under a read voltage of 0.05 V.

When irradiated by X-ray at 2 s, the image captured under a dose rate of 1.58 $\mu\text{Gy s}^{-1}$ reflects a superposition of a transient and a persistent signal (G_t and G_p), corresponding to the direct detection and dose-dependent memory modes, respectively. After the X-ray was turned off (at 2.9 s), the transient component decays, and only the persistent signal remains. This large discrepancy in the temporal response of two channels permits the unambiguous delineation of these components, as highlighted by green and red columns in Figures 4c and S17. In this way, the respective G_t and G_p images can be reconstructed to better display the information from two different channels. Further exposure to X-ray pulses gradually enhanced the image contrast till 20 s, and a clear non-volatile image of the letter “L” was obtained. This stored image can be erased by applying a reverse bias. At a lower dose rate of 0.15 $\mu\text{Gy s}^{-1}$, the device exhibited only a transient X-ray response and fully returned to its initial state once the irradiation ceased, without exhibiting any discernible memory behavior. This dose-dependent imaging response enables the preprocessing of X-ray information: weak X-ray signals can be filtered out by the G_p detection channel, which results in image contrast enhancement and background noise reduction.

The long-term memory characteristics of the device are shown in Figure 4d. The device was stored in ambient air at room temperature (25 °C) and relative humidity (55%). Stable conductance retention is observed for both saturated and non-saturated memory states over a duration of 500 min, demonstrating reliable non-volatile storage within the experimentally accessible time window. A longer retention time is promised when stored in a N_2 atmosphere (Figure S18). To substantiate the reversibility and controllability of the multilevel operation, 20 X-ray pulses ($T = 2$ s, dose rate = 2.12 $\mu\text{Gy s}^{-1}$, pulse width = 0.2 s) were applied to get potentiation with the gradual increase of device conductance from 2.7 to 38 nS. Similarly, 80 reverse voltage pulses ($T = 2$ s, -0.5 V, pulse width

= 0.2 s) encompassed the depression behavior to stepwise regain the low conductance state. As shown in Figure 4e, we demonstrated the long-term synaptic stability of perovskite devices by executing repetitive transitions between HRS and LRS by applying 1000 pulses (10 cycles). Reduction of the dose rate to 1.05 $\mu\text{Gy s}^{-1}$ yielded stable cyclic output yet a delayed saturation in conductance evolution (Figure 4f). The conductance increased from 2.5 to 37 nS and ultimately reset to 3.5 nS upon the X-ray and voltage pulses. However, we observed an asymmetry between the increases and decreases in persistent conductance, a memristive event similar to that of the biological synapses. This phenomenon should originate from the unique defect formation kinetics upon the X-ray-induced doping process.

In summary, we demonstrate an ion–electronic-defined perovskite device that enables multimodal functions from detection and memorization to image intensification of X-ray signals. Unlike traditional semiconductor detectors, the signal of such a device is distinguished by two channels from the photoelectric and radiation-doping mechanisms. Both dynamics rapidly respond to X-ray pulses, and therefore the resulting X-ray-induced conductance can be decoupled into non-volatile ionic and volatile electronic components, which correspond to the accumulated dose and real-time dose rate, respectively. We show that the device can not only achieve an X-ray sensitivity of 1159 $\mu\text{C Gy}^{-1} \text{cm}^{-2}$ and a low detection limit of 27.9 nGy s^{-1} but also allow multiple functions of the persistent photoconductivity, multi-state non-volatile storage, and super-linear dependence on radiation intensity. Compared with traditional detection systems, the in-sensor radiation detection concept in this work provides a roadmap toward a multimodal X-ray detection system that does not require any data transportation between external storage and computing components and may serve in the development of portable all-in-one devices for X-ray-based machine vision and biomedical applications.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsmaterialslett.6c00799>.

Experimental methods; XRD pattern of the as-fabricated MAPbI_3 film (Figure S1); top-view and cross-sectional SEM images of the perovskite device (Figure S2); absorption coefficients of different materials as a function of X-ray photon energy (Figure S3); bond energies and absorption coefficients for 40 keV X-ray photons of different materials (Figure S4); current–voltage (I – V) characteristics of the perovskite device measured with and without X-ray irradiation (Figure S5); switching endurance test of 100 cycles of the perovskite device under X-ray-assisted SET and electrical RESET processes (Figure S6); cumulative distribution of resistance values of LRS and HRS with X-ray-assisted SET and electrical RESET processes across 20 independent devices (Figure S7); dependence of threshold voltage and resistance state of the perovskite device under varying X-ray dose rates (Figure S8); time-dependent evolution of device conductance (Figure S9); variation of the persistent conductance as a function of pulse number (Figure S10); time-dependent conductance curve of the device under another radiation pulse (Figure S11); time-dependent response of the device under low dose rates

(Figure S12); variation of the persistent conductance as a function of accumulated dose (Figure S13); AFM images of the perovskite films after exposure to different X-ray radiation doses (Figure S14); steady-state PL spectra and the integral PL peak area ratio of the perovskite device after exposure to different radiation doses (Figure S15); TRPL decay profiles of pristine, irradiated, and reset perovskite films (Figure S16); two different signal channels (G_t and G_p) read at different pulse-on times under a 0.05 V read bias (Figure S17); and non-volatile conductance of the devices stored in a N atmosphere (Figure S18) (PDF)

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Y.L. designed and conducted the experiments. C.Z. helped with device performance measurement. X.Z. carried out the theoretical simulation. Y.L. and S.Y. wrote the manuscript. D.L. and Z.W. assisted with manuscript revision. H.G.Y., Y.H., and S.Y. provided all support needed in this work. All authors contributed to the general discussion and reviewed the paper.

Notes

The authors declare no competing financial interest.

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