

## RESEARCH ARTICLE

# Tellurium Sub-Oxides Infrared Phototransistors for Adaptive Super-Resolution Image Reconstruction

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## ABSTRACT

Infrared (IR) detection using crystalline silicon or III-V compounds is commonly utilized but often challenged by bulkiness and inefficiency. With the development of autonomous driving and machine vision, there is a growing need for IR technology to incorporate compact neural architectures. In this study, IR-sensitive *p*-type disordered tellurium sub-oxides (TeO<sub>x</sub>) thin films are deposited via an inorganic blending strategy. By integrating a luminescent dielectric layer, synergistic charge transfer and photon-induced secondary excitation endow TeO<sub>x</sub>-based IR-visible adaptive sensors (IVAS) with broadband detection and memory capabilities. The IR-driven modulation of IVAS convolutional weights enables super-resolution image reconstruction even under suboptimal conditions. This IVAS-based system achieves a peak signal-to-noise ratio of 27.55 dB (compared to 26.85 dB conventionally), a structural similarity index measure of 0.94 (compared to 0.88 conventionally), and a 13.8% reduction in mean absolute error. These findings highlight TeO<sub>x</sub>-based IVAS as a robust and adaptive solution for IR machine vision systems.

## 1 | Introduction

Current infrared (IR) detection technologies predominantly utilize photodiodes and phototransistors based on crystalline silicon or III-V compound semiconductors [1–3]. These well-established platforms are known for their high sensitivity and rapid response speeds, making them suitable for passive thermal imaging, stable performance under variable lighting conditions, and effective operation in optically challenging environments [4–7]. Despite their widespread use, these technologies face inherent limitations, particularly in the advanced fields, such as autonomous sensing, immersive visual computing, and spectrally adaptive machine vision [8]. Notably, their narrow spectral response, dependence on complex growth processes, and limited

compatibility with standard silicon-based electronics present significant barriers to large-scale integration, on-chip processing, and the development of next-generation intelligent sensing systems [9–11].

To address these limitations, alternative materials with broadband IR sensitivity, simplified processing, and compatibility with silicon-based technologies are being actively explored [6, 12]. Among these, thin-film oxide semiconductors have attracted considerable interest due to their low-temperature processability, environmental stability, and suitability for large-area processing [13]. However, most oxide semiconductors have wide bandgaps (> 2 eV), limiting their IR photoresponse [14–17]. In this context, *p*-type tellurium oxide (TeO<sub>x</sub>) emerges as a promising candidate. Its

tunable sub-1 eV optical bandgap, resulting from mixed oxidation states and disordered lattice structures, provides intrinsic IR sensitivity while maintaining the thermal and chemical stability [18, 19]. Furthermore,  $\text{TeO}_x$  is compatible with scalable, room-temperature vacuum deposition techniques, such as sputtering or thermal evaporation, enabling cost-effective and CMOS-compatible device fabrication [20, 21]. While recent advances have demonstrated high-performance *p*-type transistors based on structural engineering of  $\text{TeO}_x$  and broadband photodetection through heterojunction design, these approaches remain fundamentally confined to the conventional paradigm of single-mode photodetection or electronic switching [22, 23]. Furthermore, the development of  $\text{TeO}_x$ -based systems is hindered not only by an incomplete understanding of composition–structure–property relationships in disordered phases but also by the lack of architectures capable of integrating sensing, memory, and computational functions. Critically, the potential of  $\text{TeO}_x$  for constructing adaptive perception systems remains largely unexplored, leaving a significant gap between its device performance and applicability in intelligent machine vision.

In this work, we employ an inorganic blending strategy to fabricate *p*-type disordered  $\text{TeO}_x$  thin films, achieving intrinsic IR sensitivity and tunable electronic properties. Apart from material optimization, we have constructed an infrared-visible adaptive sensor (IVAS) through the integration of a luminescent dielectric layer, where synergistic charge transfer and cascaded secondary excitation enable broadband detection and memory functionality. Most significantly, the IR illumination serves as a dynamic control signal, enabling adaptive weight modulation for processing visible-light images. By directly converting IR signals into conductance changes, it implements a light control mechanism. This approach fundamentally simplifies the system architecture by merging the control and computing phases, thereby facilitating in-situ weight modulation. The efficacy of this IR-driven adaptation is validated through robust super-resolution (SR) image reconstruction under challenging lighting conditions. With IR-driven adaptive weight optimization enabled by IVAS, we achieve performance improvements, including a peak signal-to-noise ratio (PSNR) of 27.55 dB (compared to 26.85 dB conventionally), a structural similarity index measure (SSIM) of 0.94 (vs 0.88 conventionally), and a 13.8% reduction in mean absolute error (MAE).

## 2 | Results and Discussion

### 2.1 | SR Reconstruction Enabled by IVAS Array

To address the growing demands for adaptive sensing, we introduce an IVAS architecture that couples a *p*-type  $\text{TeO}_x$  thin-film with a luminescent dielectric layer. As shown in Figure 1a, the IVAS exhibits a dual-mode photoresponse, enabling broadband sensitivity across the visible and IR region. This wide spectral response arises from the synergistic interplay between interfacial charge transfer and photon-induced secondary excitation (Figure 1b). After that, the IVAS array is directly embedded into a super-resolution convolutional neural network (SRCNN) framework, functioning not only as a photodetector but also as a hardware-level memory unit (Figure 1c). Light-induced conductance modulation in the  $\text{TeO}_x$  channel can be retained

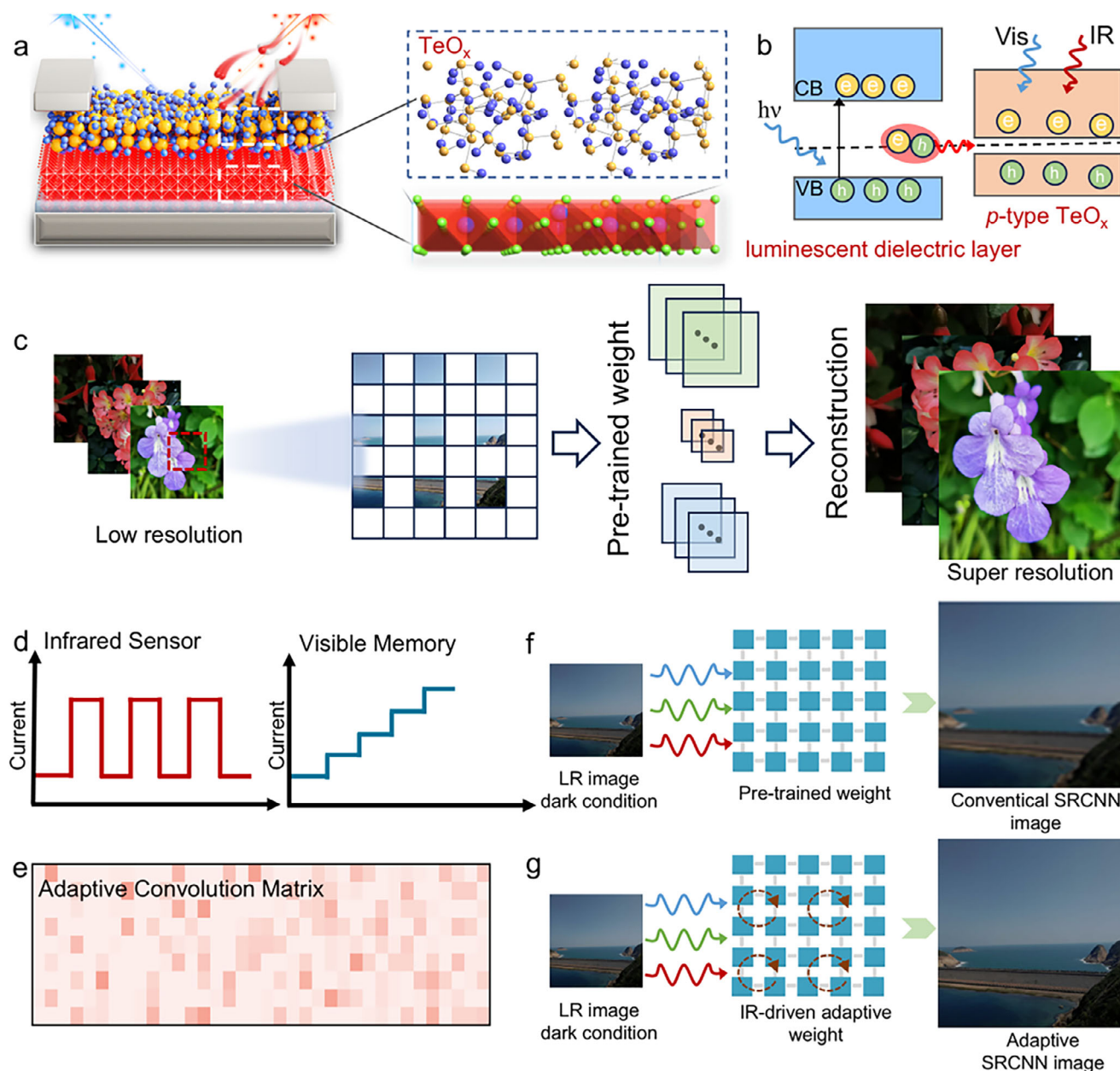
and mapped to convolutional weights. Each IVAS pixel operates as a reconfigurable unit under IR illumination, where the corresponding conductance programming supports environmental adaptation (Figure 1d). This dynamic IR-driven weight modulation facilitates the network to adapt its parameters in response to challenging lighting (Figure 1e). As demonstrated in Figure 1f, conventional SRCNN fails to reconstruct image fidelity under low-light conditions [24]. In contrast, the IVAS-integrated system recovers key spatial features by leveraging IR-driven weight optimization (Figure 1g). This highlights the potential of IVAS as a foundational element in intelligent, environment-adaptive machine vision platforms.

### 2.2 | Characterizations of *p*-Type Disordered $\text{TeO}_x$ Films

The IVAS device was fabricated using a *p*-type  $\text{TeO}_x$  channel and a luminescent dielectric layer, as confirmed by high-resolution scanning transmission electron microscopy (HR-STEM) and energy-dispersive X-ray spectroscopy (EDS) elemental mapping (Figure 2a). The  $\text{TeO}_x$  thin-film with a thickness of  $\sim 8$  nm was deposited via room-temperature thermal evaporation (See the Methods section). The uniform and continuous  $\text{TeO}_x$  films were obtained using an inorganic blending strategy, exhibiting a smooth surface with a roughness of 0.32 nm (Figures S1 and S2). As shown in Figure 2b, it reveals that the  $\text{CsPbI}_3$  perovskite quantum dots (PQDs) display distinct diffraction spots. In contrast, the  $\text{TeO}_x$  layer presents diffuse diffraction rings, confirming its disordered property. The characteristic vibrational modes of the helical chain structure in our  $\text{TeO}_x$  are revealed by Raman analysis, which shows three well-resolved peaks assigned to fundamental phonon modes (Figure 2c) [25]. Furthermore, X-ray photoelectron spectroscopy (XPS) verifies the simultaneous presence of both elemental  $\text{Te}^0$  and oxidized  $\text{Te}^{4+}$  states (Figure 2d) [26–28].

During thermal evaporation, the oxygen content of  $\text{TeO}_x$  films can be precisely tuned by adjusting the  $\text{Te}/\text{TeO}_2$  source ratio (Figure S3), enabling systematic investigation of structural characteristics across varying oxygen stoichiometries ( $x = 1.12, 1.60$ , and 2). X-ray diffraction (XRD) patterns of all compositions show the absence of diffraction peaks (Figure 2e; Figure S4), indicating that they maintain their disordered properties [20, 29, 30]. To further elucidate the amorphization mechanism, melt-quenching-based molecular dynamics (MD) simulations were performed (Figure 2f; Figure S5) [31, 32]. Radial distribution function (RDF) analysis reveals the retention of short-range  $\text{Te}-\text{O}$  bonding ( $\sim 2$  Å) in disordered  $\text{TeO}_x$ . At the same time, long-range order characteristic of the crystalline phase is suppressed (Figure 2g; Figure S6). These findings suggest that the structure preserves local coordination motifs and that disordering predominantly disrupts extended atomic networks [33, 34].

To elucidate the oxidation-state-dependent charge transport modulation, we conducted electrical characterization on  $\text{TeO}_x$  films ( $x = 1.12$  and 1.60). The bare  $\text{TeO}_x$  ( $x = 1.12$ )-based transistor exhibited a field-effect mobility ( $\mu_{\text{FE}}$ ) of  $3.80 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$ , while the  $\text{TeO}_x$  ( $x = 1.60$ ) device showed a reduced  $\mu_{\text{FE}}$  of  $1.52 \text{ cm}^2 \cdot \text{V}^{-1} \cdot \text{s}^{-1}$  (Figure S7). This pronounced mobility disparity results from the introduction of higher oxygen levels. This process creates a



**FIGURE 1** | (a) Structural schematic of the IVAS device, highlighting its broadband photodetection under visible and IR illumination. (b) Operational mechanism showing synergistic charge transfer and photon-induced secondary excitation. (c) Neural network architecture of SRCNN. (d) Dual-mode functionality of IVAS, enabling IR sensing and visible-light memory. (e) Adaptive weight mapping protocol within the SRCNN enabled by IR-driven modulation. (f) Conventional super-resolution under dark conditions (g) Enhanced image reconstruction achieved through IR-driven adaptive weight modulation in SRCNN. Photograph credits of 1c, 1f, and 1g: He Shao.

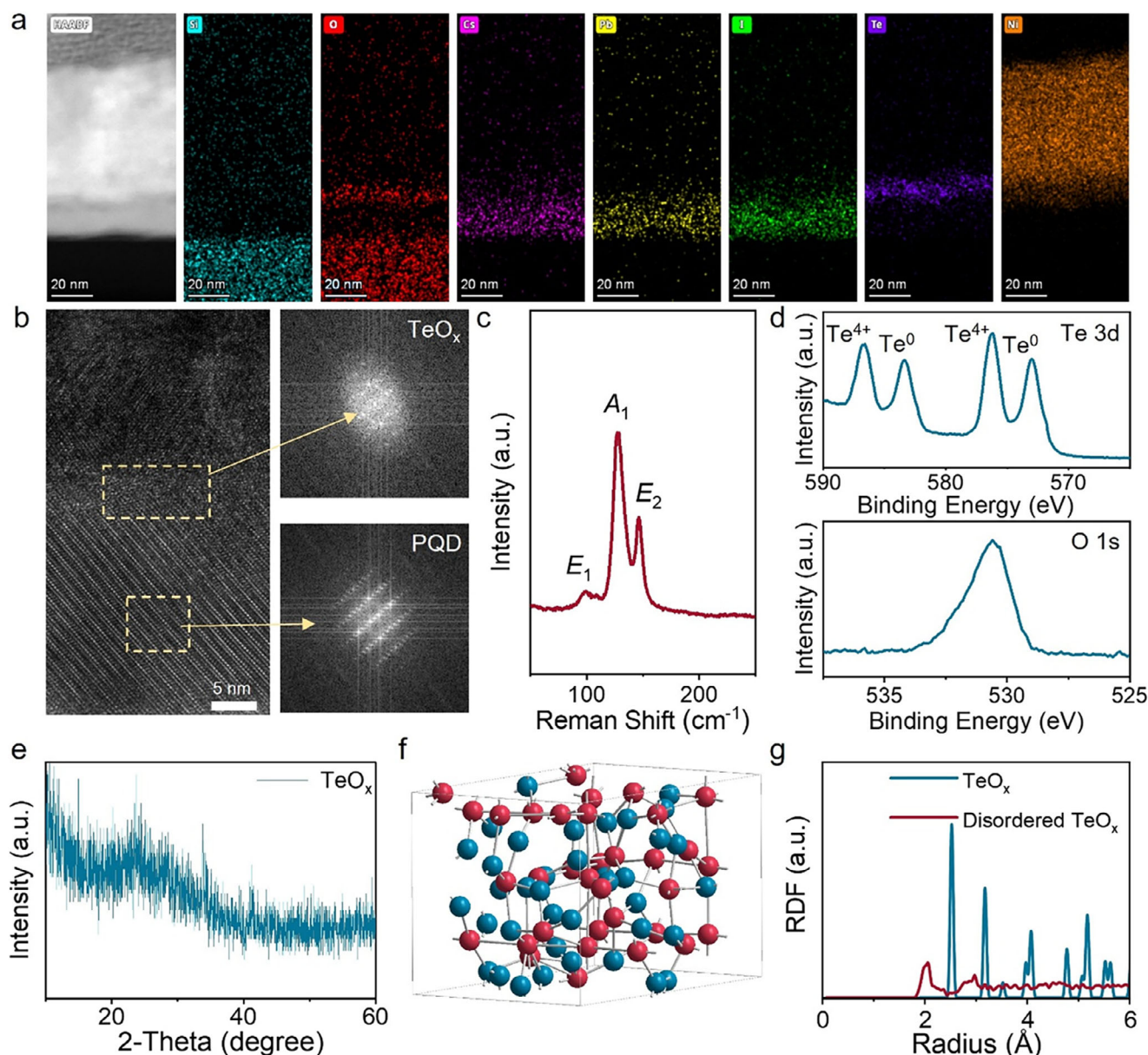
dense distribution of trap states and scattering centers, which in turn impede carrier transport [35, 36]. Despite its lower mobility, the  $\text{TeO}_x$  ( $x = 1.60$ ) device displays an enhanced  $I_{\text{on}}/I_{\text{off}}$  ratio, attributable to improved carrier regulation at a cost of the reduced charge percolation [37, 38].

### 2.3 | Luminescent Dielectric Engineering of IVAS

After identifying  $\text{TeO}_x$  with balanced charge transport and switching characteristics, the  $\text{TeO}_x$ -based IVAS was subsequently fabricated by integrating with a luminescent dielectric layer. The

luminescent dielectric layer consists of  $\text{CsPbI}_3$  PQDs embedded in polystyrene (PS). The PQDs were synthesized via the hot-injection method (as illustrated in Figure S8), yielding a uniform size distribution and stable black-phase characteristics (Figures S9 and S10) [39, 40]. The luminescent dielectric layer mainly contributes to enhanced optoelectronic performance in visible regions while preserving the fundamental electronic characteristics of  $\text{TeO}_x$  films. As shown in Figures S11 and S12, the transfer and output characteristics exhibit typical *p*-type behavior, with an on/off ratio of  $10^4$  and a mobility of  $2.5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ . Furthermore, the influence of  $\text{TeO}_x$  film thickness on device behavior was explored (Figure S13), with statistical mobility and





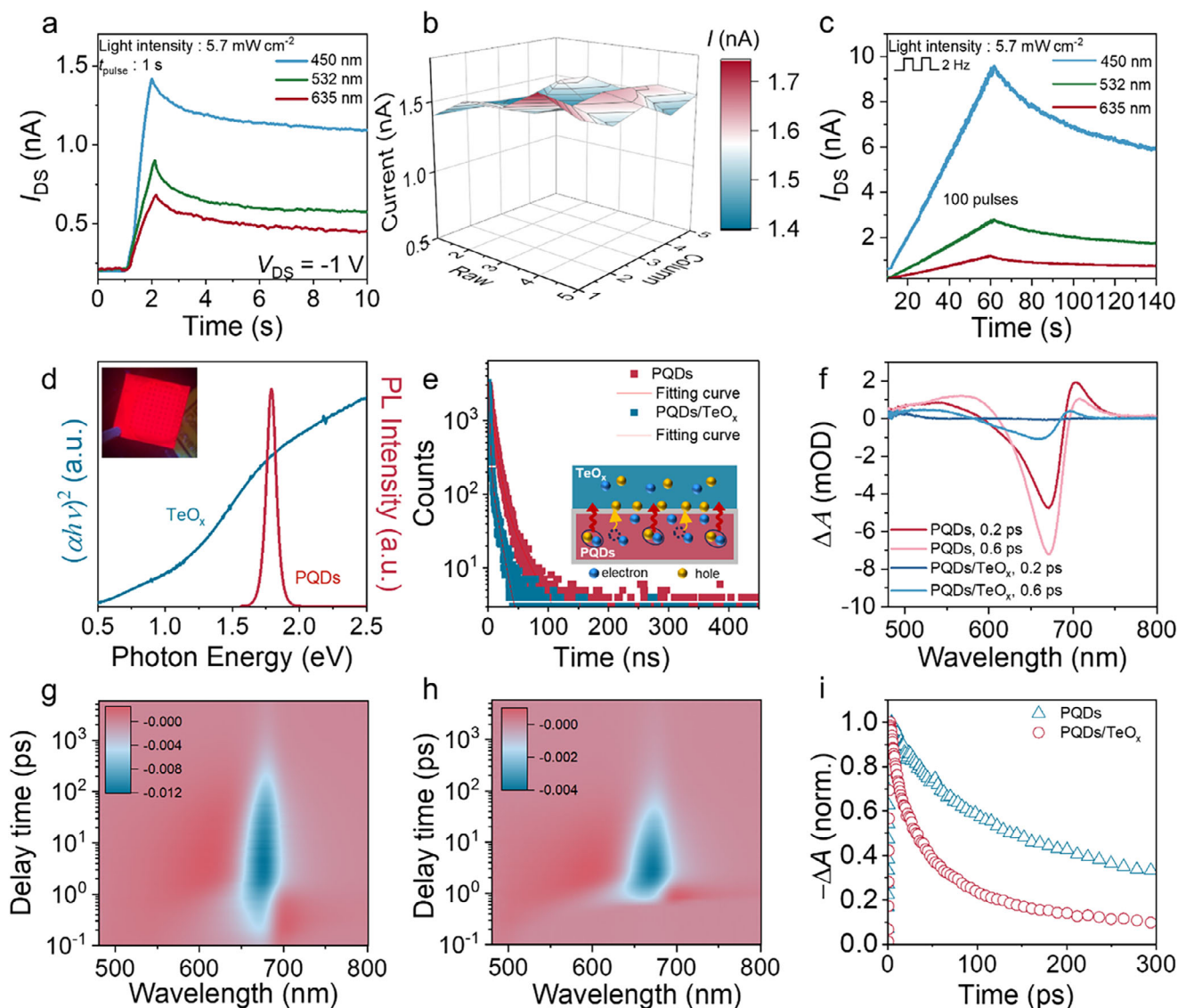
**FIGURE 2** | (a) Cross-sectional HR-STEM image and corresponding EDS elemental mapping of the device. (b) HRTEM image showing the layered device architecture. (c) Raman spectra of the TeO<sub>x</sub> film. (d) XPS of Te 3d and O 1s core levels of the TeO<sub>x</sub> film. (e) The XRD pattern of the disordered TeO<sub>x</sub> film. (f) Atomic structure of the disordered TeO<sub>x</sub>. (g) RDFs of the disordered and crystalline TeO<sub>x</sub>.

$I_{\text{on}}/I_{\text{off}}$  summarized in Figure S14. These findings reveal that increasing the TeO<sub>x</sub> film thickness beyond 10 nm results in higher off-state currents and a reduced on/off current ratio, due to the presence of excessive unoxidized Te [41]. Conversely, decreasing the thickness below 6 nm results in a significant reduction in mobility, primarily due to an insufficient carrier density.

To assess device uniformity and operational consistency, a 5 × 5 IVAS array was fabricated and characterized (Figure S15). Individual devices exhibited intense and persistent photocurrent responses under illumination at 450, 532, and 635 nm (Figure 3a) [42]. The current mapping across the array (Figure 3b; Figure S16) reveals uniform responsivity, confirming the reproducibility of the fabrication process. Under 100 consecutive light pulses (2 Hz), devices display robust and stable memory behavior across all tested wavelengths (Figure 3c), highlighting their capabil-

ity for broadband optical memory. The memory mechanism in our devices is governed by complementary charge-trapping processes. The trapping state at the TeO<sub>x</sub>/PQDs heterointerface enables the persistent photocurrent, while intrinsic defects within the PQDs and the surrounding PS matrix further contribute to charge retention [43–45]. In contrast, control devices comprising only TeO<sub>x</sub> channels exhibited volatile behavior under identical conditions (Figure S17), underscoring the essential role of the dielectric layer in enabling persistent photocurrent memory.

To further elucidate the synergistic optical interactions within the hybrid structure, a cascaded secondary excitation mechanism is proposed and experimentally validated. As shown in Figure 3d, the photoluminescence (PL) spectrum of the luminescent layer exhibits a sharp emission peak centered at ~685 nm, with the inset



**FIGURE 3** | (a) Photoresponse of IVAS under a single light pulse at 450, 532, and 635 nm ( $V_{DS} = -1$  V and  $V_R = 5$  V). (b) Spatial photocurrent distribution across the 5 × 5 IVAS array under 450 nm illumination. (c) Wavelength-dependent response under 100 consecutive light pulses. (d) Absorption spectrum of the TeO<sub>x</sub> film and PL spectrum of the PQD film. Inset: IVAS array under 365 nm UV illumination. (e) TRPL of the PQD film and PQDs/TeO<sub>x</sub> composite film. (f) TA of the PQD film and PQDs/TeO<sub>x</sub> composite film at different delay times. The TA contour maps ( $\Delta A$ - $\lambda$ -time) of the (g) PQD film and (h) PQDs/TeO<sub>x</sub> composite film under 450 nm excitation. (i) Decay dynamics at 670 nm ( $\lambda_{ex} = 450$  nm).

photograph displaying red luminescence under optical excitation [46]. Concurrently, the TeO<sub>x</sub> layer demonstrates strong optical absorption in the near-IR region, attributed to its narrow bandgap of approximately 0.8 eV. Critically, the emission spectra of the luminescent layer overlap substantially with the absorption edge of TeO<sub>x</sub>, enabling the emitted photons to be reabsorbed by the TeO<sub>x</sub> channel. This reabsorption triggers a secondary photoexcitation process within TeO<sub>x</sub>. Thus, the observed memory behavior is also primarily governed by the synergistic interplay between cascaded photon recycling and efficient charge transfer at the heterointerface. Although alternative pathways such as trap-assisted tunneling and interface dipole effects were considered, the collective evidence most consistently supports this coupled mechanism as being responsible for the memory behavior.

Following this, PL and time-resolved PL (TRPL) measurements were conducted to investigate the charge transfer behavior. As shown in Figure S18, the TeO<sub>x</sub>/PQDs composite film exhibits a significant reduction in photoluminescence (PL) intensity compared to the pristine PQD film. This fluorescence quenching is corroborated by the TRPL results in Figure 3e, which show a markedly shortened lifetime in the composite film [47, 48]. The observed simultaneous decrease in both PL intensity and lifetime provides strong evidence for the efficient charge transfer from the PQDs to the TeO<sub>x</sub> layer [45, 49]. To gain further insights into the excited-state dynamics, femtosecond transient absorption (TA) spectroscopy was performed on both pristine and composite films under 450 nm excitation. The TA spectra at 0.2 and 0.6 ps delay (Figure 3f) show a pronounced ground-state bleaching

(GSB) centered near 670 nm, corresponding to the PQDs band edge. In pristine PQDs, a positive excited-state absorption (ESA) component emerges beyond 685 nm, which is notably suppressed in the PQDs/TeO<sub>x</sub> composite structure.

The two-dimensional TA contour mappings (Figure 3g,h) further visualize the temporal evolution of this photo-induced evolution. Compared to the PQD film, the composite film shows an accelerated decay of both GSB and ESA signals, indicating faster carrier relaxation dynamics [50, 51]. The suppression of ESA in the composite film is particularly significant in the red region, implying that the emitted photons are reabsorbed by the broadband-absorbing TeO<sub>x</sub> layer. To quantify the relaxation kinetics, the GSB decay traces at 670 nm were extracted (Figure 3i). The composite film exhibits a much shorter carrier lifetime than the pristine PQDs, in agreement with the TRPL results. The consistent trend in TRPL and TA confirms that the TeO<sub>x</sub> layer serves as an energy extraction channel through radiative reabsorption.

## 2.4 | IR-Visible Synergistic Effect in IVAS Array

Beyond the visible light irradiation, the IVAS device exhibits pronounced sensitivity to IR illumination. As shown in Figure 4a,b, the photocurrent increases monotonically with increasing light intensity, highlighting the device's broadband and tunable optoelectronic response. The observation of a pronounced photoreponse in a bare TeO<sub>x</sub>-transistor under 1550 nm irradiation confirms that the TeO<sub>x</sub> layer itself is the primary site for IR detection (Figure S19). This is consistent with its intrinsically narrow bandgap (~0.8 eV) and pronounced Urbach tail, which provides localized states for efficient photon capture via phonon-assisted transitions. Furthermore, the precise voltage-dependent photocurrent modulation in the IVAS (Figure 4c) substantiates that direct photocarrier generation and control occur within the TeO<sub>x</sub> layer, facilitating efficient photocurrent generation [52, 53].

To investigate the wavelength-selective modulation, sequential visible and IR irradiation were applied to the device (Figure S20). As shown in Figure 4d, under continuous 450 nm optical pulses at 0.1 Hz, the device exhibits distinct multi-level conductance states. During the intervals between pulses, stimulation with higher-frequency IR light induces discrete conductance levels. As shown in Figure 4e, a total of sixteen conductance states (4 bits) are uniformly distributed and exhibit an approximately linear relationship, which is critical for analog computing and dynamic weight mapping in neural networks. A statistical analysis of conductance modulation ratios under IR and visible illumination ( $\sigma/\mu$ ) reveals a pronounced initial-state-dependent response, suggesting a synergistic and programmable conductance tuning operation (Figure 4f). This dynamic persists under additional visible-light wavelengths (Figure S21), further validating the broadband feedback process.

The underlying mechanisms of IVAS under illumination are further clarified through an analysis of the energy band structure (Figure 4g,h). Absorption and UPS measurements (Figures S22 and S23) confirm the formation of a type-I heterojunction between PQDs and TeO<sub>x</sub>, promoting directional carrier transport

[54]. Upon visible-light exposure, both PQDs and TeO<sub>x</sub> undergo excitonic dissociation, resulting in the generation of electron-hole pairs. The photogenerated holes in PQDs migrate into the TeO<sub>x</sub> layer, while electrons remain trapped within the PQDs or at the interface, resulting in persistent charge accumulation and a memory effect. Moreover, photoluminescent emission from PQDs provides an additional energy recycling pathway, wherein the emitted red photons are reabsorbed by TeO<sub>x</sub> through cascaded photon harvesting. Under 1550 nm illumination, photogenerated carriers in TeO<sub>x</sub> exhibit rapid recombination, leading to a characteristic volatile photocurrent behavior. The synergy effect of visible-induced charge retention and IR-induced volatile photoresponse provides an adaptive optoelectronic platform.

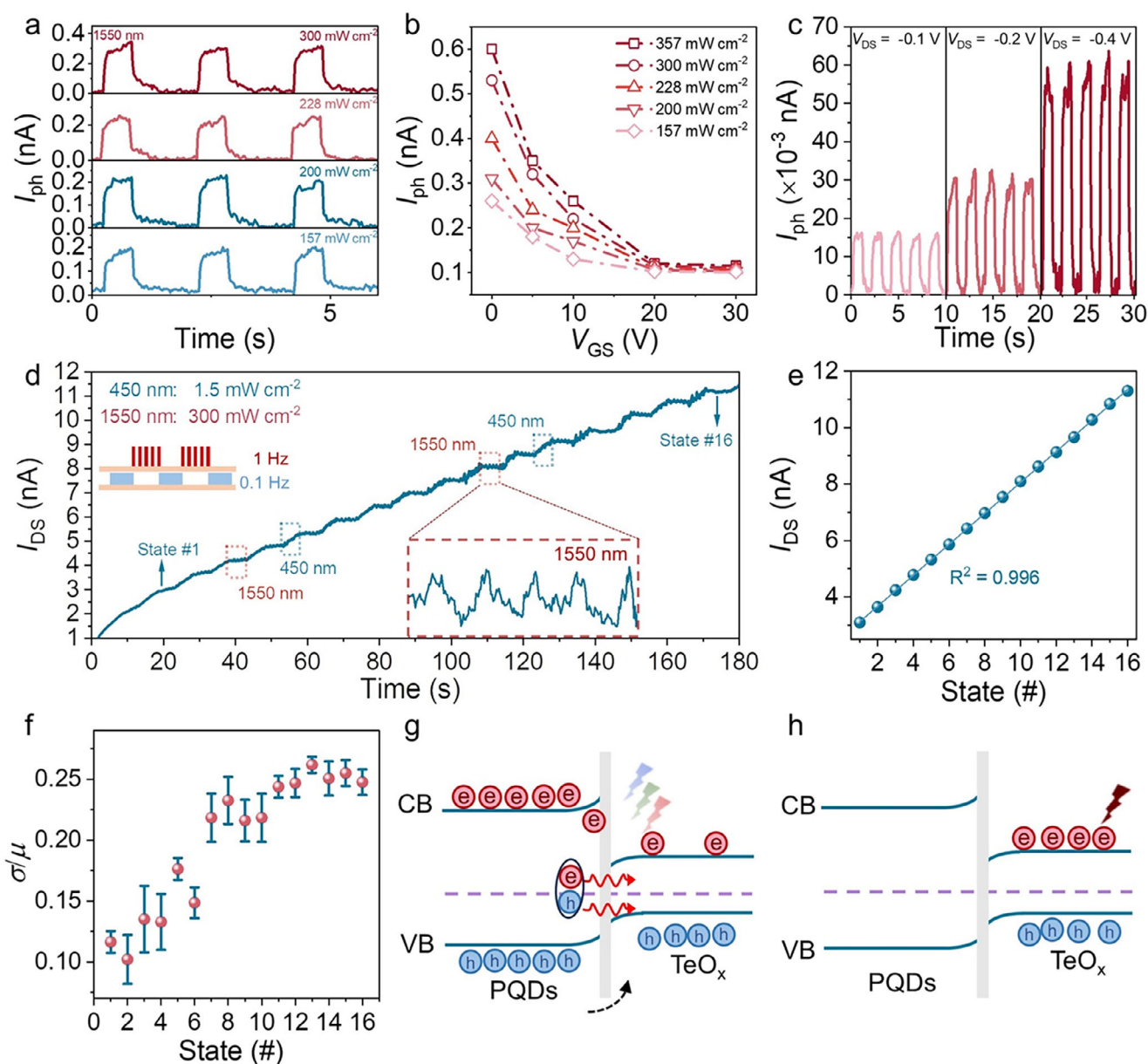
## 2.5 | IR-Driven Adaptive Image SR Reconstruction

The IVAS architecture was further integrated with SRCNN to achieve simultaneous high-fidelity image reconstruction and enhanced feature extraction (Figure 5a) [55, 56]. The SRCNN framework was specifically selected for its architectural compatibility with our hardware implementation, where its constrained network depth and convolutional topology significantly relax the precision requirements for conductance control in the array (Figure S24). The super-resolution performance of the IVAS architecture was evaluated using a DIV2K dataset for training, with low-resolution inputs prepared for  $2 \times$  upscaling [24]. After training the SRCNN to obtain optimized weights, the learned parameters were mapped to the conductance states of our hardware array. Experimentally captured images were then used for testing. A systematic comparison was conducted between the software-based model and the IVAS-implemented counterpart to assess their respective reconstruction performance. As shown in Figure 5b, both software-based and IVAS-implemented SRCNNs achieve PSNR values exceeding 28 dB based on the pre-trained weights, confirming the preservation of structural integrity and detail.

To systematically evaluate the robustness of the IVAS architecture to luminance variations in super-resolution tasks, we conducted comparative experiments under both standard and reduced brightness conditions. The original images were processed to create a controlled low-light benchmark, with brightness reduced by 40% to simulate challenging imaging conditions (Figure 5c; Note S1). An SRCNN model was first pre-trained on normal-light images to obtain baseline weights (Figure 5d; Notes S2 and S3). Crucially, instead of creating multiple specialized models, we trained a unified system. It was achieved by jointly optimizing the baseline weights with a modulation network that learns to generate weight perturbations ( $\Delta W$ ) in response to IR illumination. The modulated weights  $W_{\text{effective}} = W_{\text{base}} + \alpha \Delta W_{\text{IR}}$  are optimal for the current lighting condition. These modulated weights are integrated into all three convolutional layers of the SRCNN (Note S4), enabling dynamic feature extraction.

As illustrated in Figure 5e, for inputs with normal brightness, SRCNN achieves higher reconstruction quality compared to bicubic interpolation, particularly in edge clarity and detail retention. The PSNR values are confirmed to be 27.65 dB (bicubic) and 28.40 dB (SRCNN). Under low-light conditions,



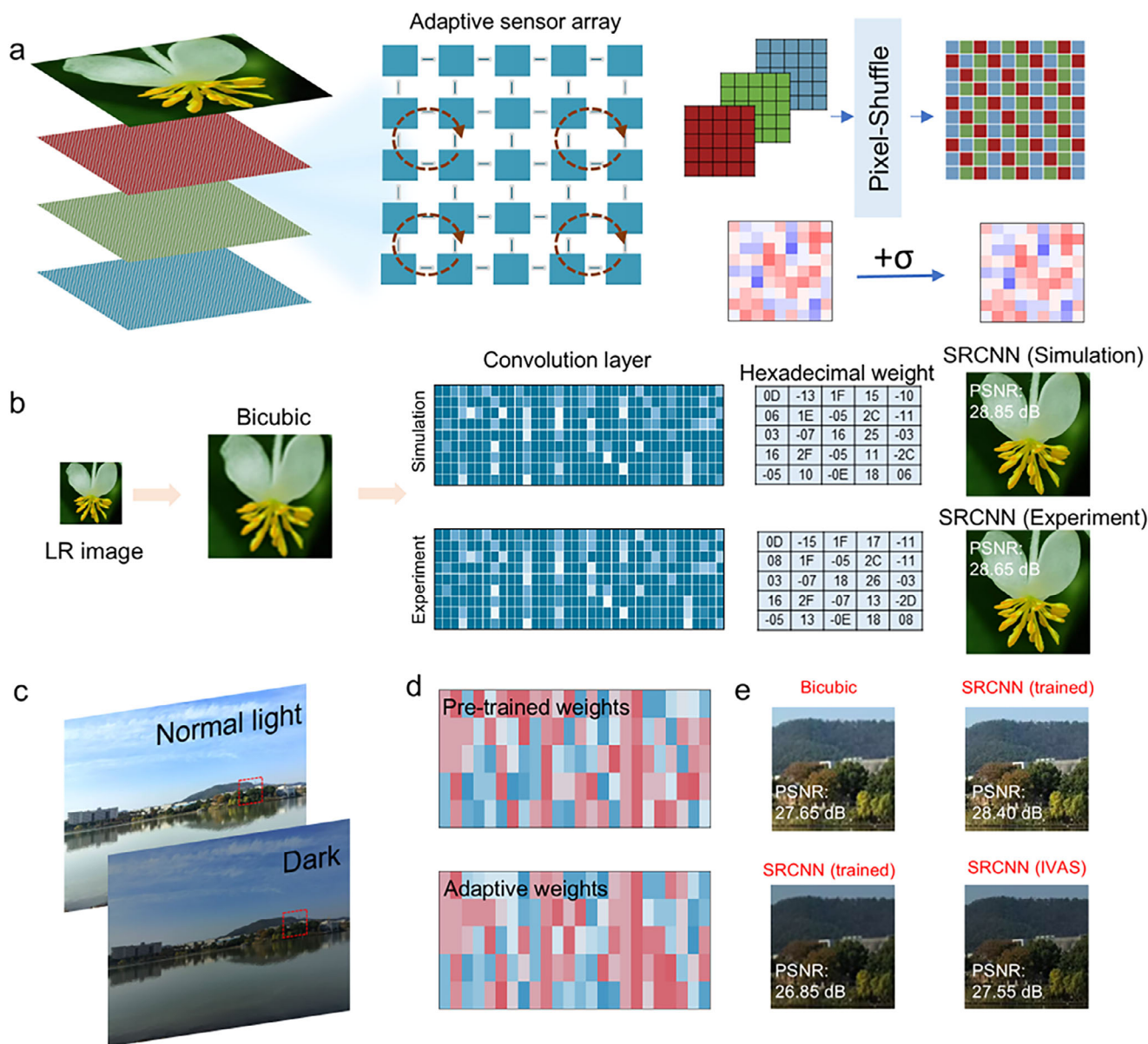


**FIGURE 4** | (a) Photoresponse under 0.5 Hz switching with varying 1550 nm intensities ( $V_{DS} = -1$  V and  $V_R = 5$  V). (b) Photocurrent of IVAS under 1550 nm light illumination as a function of gate voltage and light intensity. (c) Photoresponse with varying  $V_{DS}$ . (d) Multi-level conductance tuning by 450 nm pulses and 1550 nm pulses. (e) Linear fit between conductance state and pulse number. (f) Statistical analysis of photoconductance changes ( $\sigma$ : 1550 nm-induced variation;  $\mu$ : per-pulse change for 450 nm) across sixteen conductance states. (g,h) Energy band evolution and charge carrier transfer process of IVAS under visible and IR illumination.

the conventional SRCNN with pre-trained weights exhibits significant performance degradation. In contrast, the fine-tuned model demonstrates marked improvements, benefiting from the IR-assisted functionality of the IVAS array. The PSNR improves from 26.85 to 27.55 dB, SSIM increases from 0.88 to 0.94, and MAE is reduced by 13.8%, indicating the array's ability to recover features in poorly illuminated scenes. Furthermore, the IVAS architecture leverages local conductance adaptation in different brightness environments to mitigate pixel saturation effects. As shown in Figure S25, these improvements highlight the array's capacity for preserving image details under undesirable lighting conditions.

### 3 | Conclusion

The proposed optoelectronic IVAS array, featuring IR-assisted adaptive weight modulation, offers a revolutionary approach to achieving efficient super-resolution imaging. By leveraging a *p*-type TeO<sub>x</sub> channel layer coupled with a luminescent dielectric layer, the IVAS system dynamically adjusts convolutional weights to ensure robust feature extraction under diverse lighting conditions. The integration of IR modulation significantly enhances image reconstruction quality, as evidenced by an increase in PSNR from 26.85 to 27.55 dB, an improvement in SSIM from 0.88 to 0.94, and a 13.8% reduction in MAE compared to conventional



**FIGURE 5** | (a) Schematic of the multimodal processing strategy of the IVAS array. (b) Performance comparison of HR image reconstruction between software-based and IVAS-based SRCNN. (c) Scene images under normal brightness and 40% brightness reduction. (d) Pre-trained SR weights (normal brightness) versus IR-driven weights under low-light conditions. (e) Bicubic versus SRCNN on normal-brightness images (top) and fixed pretrained weights versus adaptive weights on low-brightness images (bottom). Photograph credits of 5a, 5b, 5c, and 5e: He Shao.

SRCNN approaches. As the demand for intelligent and adaptive imaging systems increases, the IVAS approach presents a promising pathway to address these challenges, leveraging the strengths of advanced materials science and cutting-edge computational techniques.

## 4 | Experimental Section/Methods

### 4.1 | Measurement and Characterization

The optical properties of PQDs and  $\text{TeO}_x$  thin films were measured using Shimadzu UV-3600 and FLS-980 (Edinburgh Instruments). The femtosecond TAS was operated with the pump pulse of 450 nm, 150 fs, 1 kHz, and 100  $\mu\text{J}/\text{cm}^2$ . The morphologies

of the  $\text{TeO}_x$  films were confirmed using AFM (Bruker Dimension Icon AFM) and SEM (Hitachi S3400N). The crystal structure of PQDs and  $\text{TeO}_x$  films was confirmed by XRD (D2 PHASER, Bruker). The chemical bonding and elemental composition were characterized by Raman spectroscopy (WITec RAMAN alpha 300R) and XPS measurements (Escalab 220i XL, VG Scientific). The structure and EDS mapping were examined using high-resolution transmission electron microscopy (HRTEM, Thermo Scientific, Talos F200X). The optoelectronic properties of the fabricated devices were evaluated using an Agilent 4155C semiconductor parameter analyzer combined with a precision probe station. For photodetector measurements, lasers with different wavelengths, including visible (450, 532, and 635 nm) and infrared (1550 nm), were used as light sources. The incident light powers (P) were determined using a power meter (PM400, Thorlabs).



## 4.2 | Device Fabrication

Bottom-gate top-contact thin-film transistors were fabricated on heavily doped *p*-type silicon substrates with 270 nm of thermally grown SiO<sub>2</sub>. The fabrication process involved the following sequential steps: First, the SiO<sub>2</sub> surface was subjected to oxygen plasma treatment (5 min, 50 W) to enhance surface hydrophilicity and remove organic contaminants. Subsequently, 10 mg/mL PQDs solution in toluene was mixed with 6 mg/mL PS solution at a 1:1 volume ratio. The mixture was then spin-coated onto the substrate at 3000 rpm for 60 s. The synthesis process of CsPbI<sub>3</sub> PQDs was introduced in Figure S8. The deposition sources with controlled oxygen stoichiometry were synthesized through solid-state reactions between high-purity Te (99.999%) and TeO<sub>2</sub> (99.99%) powders, where the TeO<sub>x</sub> (*x* = 1.12) composition was achieved using a 270:30 mg (9:1 molar ratio) Te/TeO<sub>2</sub> mixture and the TeO<sub>x</sub> (*x* = 1.60) composition was prepared with a 250:50 mg (5:1 molar ratio) Te/TeO<sub>2</sub> mixture. The mixture was then deposited onto the substrate using thermal evaporation under high-vacuum conditions (base pressure < 4 × 10<sup>-6</sup> Torr) at a controlled deposition rate of 0.8 Å/s. Film thickness was precisely monitored in situ using an INFICON SQC-310 deposition controller and a quartz crystal microbalance. Subsequently, the substrate was annealed in air at 100 °C for 30 min to stabilize its oxidation state. Both the active channel and source/drain electrodes were defined through shadow mask patterning to minimize gate leakage currents and ensure accurate electrical characterization. The device fabrication was completed by electron-beam evaporation of nickel source/drain electrodes through the shadow mask.

## 4.3 | DFT Calculation

All calculations are performed within the framework of density functional theory using the projector augmented plane-wave method, as implemented in the Vienna ab initio simulation package. The generalized gradient approximation proposed by Perdew, Burke, and Ernzerhof is selected for the exchange-correlation potential. The long-range van der Waals interaction is described by the DFT-D3 approach. For electronic structure calculations, a plane-wave cutoff energy of 420 eV was employed, and the Kohn–Sham equations were solved with an energy convergence criterion of 10<sup>-5</sup> eV. Gamma-point sampling was used for the Brillouin zone integration in all calculations. The disordered amorphous TeO<sub>x</sub> (*x* = 1.12) structures were generated through a melt-quenching protocol within the framework of ab initio molecular dynamics (AIMD). The initial atomic configurations were randomized by premelting at 5000 K for 1 ps, then melting at 1200 K for 5 ps and rapid quenching to 400 K with a cooling rate of -100 K/ps. Final structural relaxations were performed until atomic forces converged to less than 0.02 eV/Å.

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### Conflicts of Interest

The authors declare no conflict of interest.

### Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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## Supporting Information

Additional supporting information can be found online in the Supporting Information section.

**Supporting file:** adma72046-sup-0001-SuppMat.docx