

Quad-Band Short-Wave Infrared Detection Using Bandgap-Controlled Ag₂Te Quantum Dots

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Multispectral photodetection in the short-wave infrared (SWIR) range has been highly desirable for applications such as night vision, quality inspection, and bio-imaging. Colloidal quantum dots (QDs) have been extensively explored as a promising alternative to conventional epitaxial semiconductors for multispectral photodetection owing to their size-tunable bandgap-controlled optoelectronic properties and potential for low-cost, high-throughput fabrication. Here, a multispectral SWIR photodetector is demonstrated based on four distinct sizes of heavy-metal-free Ag₂Te QDs, each patterned side-by-side using a direct QD photopatterning process— involving QD film deposition, ultraviolet-induced selective QD ligand crosslinking, and pattern development. This enables precise spatial integration of the four different QD channels into a single device platform that is independently addressable, allowing integrated multispectral SWIR detection within a monolithic platform. The resulting multispectral Ag₂Te QD channels show maximum responsivities of 188.4, 173.5, 155.6, and 185.8 mA W⁻¹ at 1150, 1350, 1550, and 1800 nm, respectively. By leveraging the distinct photoresponses of four spectrally differentiated Ag₂Te QD channels to SWIR input light, it is demonstrated that the multispectral photodetector enables simple, non-spectrometric wavelength identification—highlighting a unique capability unattainable with conventional single-size-QD devices.

vision,^[1] remote sensing,^[2] night vision,^[3] bio-imaging,^[4,5] industrial inspection,^[6] and environmental monitoring.^[7,8] This approach can be implemented using a series of photodetectors or optical filters that can secure specific wavelength bands.^[9,10] Such devices typically require multiple IR active materials exhibiting spectral responses at desired regions of the electromagnetic spectrum to be integrated into a single imaging device. A well-known integration strategy involves vertically stacking these materials.^[11,12] Typically, the use of a stacked multi-band structure that includes different IR active materials remains the most conventional approach to date.^[13,14] These stacked systems mainly incorporate epitaxially grown semiconductors or combinations of these semiconductors by forming type-II superlattices and quantum-well structures.^[13–17] However, these epitaxial thin films present technical challenges such as need for lattice matched substrates, high fabrication complexity and high fabrication cost, and low production yield,

limiting the widespread adoption of such technologies in civilian applications.^[18,19]

Colloidal quantum dots (QDs) have recently been applied as a promising alternative, cost-effective solution to the aforementioned IR-sensitive semiconductors, owing to the characteristic size-tunable bandgaps of QDs that allow a specific

1. Introduction

Multispectral infrared (IR) photodetection, responding to IR light with multiple spectral wavelength bands, has garnered attention owing to the potential to advance object identification by analyzing spectral information in applications such as machine

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detection wavelength to be targeted.^[20–23] Multispectral photodetectors based on QDs can be configured in two ways. More specifically, QDs with different absorption bands can either be stacked vertically or patterned side-by-side.^[24–27] For example, a dual-band QD photodetector, with differently sized HgTe QDs stacked vertically in a back-to-back configuration, has been demonstrated for sensing short-wave infrared (SWIR) and mid-wave infrared (MWIR) signals by switching the applied bias voltage.^[25] Although multispectral detection was possible using a single device structure, the devices with a back-to-back configuration can only detect two different IR signals separately under the respective switching conditions. Therefore, multispectral signals could not simultaneously be acquired. Additionally, a voltage-readout scheme to apply this structure to Si readout integrated circuitry (ROIC) has not been fully established, limiting the imaging capability of the structure. In addition, an ultrabroadband QD photodetector encompassing the visible to the MWIR range has been fabricated by stacking two different PbS and HgTe QDs to decouple visible, SWIR, and MWIR signals using respective optical filters on top.^[26] Although the ultrabroadband detection achieved multispectral imaging using optical filters, these devices typically suffer from spectral crosstalk and decreased sensitivity. Alternatively, multispectral photodetectors arranged with different QD pixels placed side-by-side were demonstrated, similar to the manner in which different colored QD patterns are pixelated side-by-side in a QD electroluminescence display.^[27–29] For example, a multispectral IR QD photodetector encompassing light signals from the ultraviolet (UV) to SWIR range was fabricated using pixels from an HgTe QD photoconductor and a down convertor containing an Si-based complementary metal oxide semiconductor (CMOS) ROIC coupled with CsPbX₃ perovskite nanocrystals.^[27] Despite this progress, the multispectral IR QD photodetectors developed thus far involve IR-sensitive materials including hazardous heavy metals, such as Pb and Hg. These materials do not comply with the Restriction of Hazardous Substances (RoHS) regulations and are strictly regulated from use in industry.^[30]

Herein, we present a multispectral SWIR QD photodetector fabricated by directly photopatterning heavy-metal-free Ag₂Te QDs with four different sizes. These QDs have recently garnered significant attention as RoHS compliant IR QDs containing environmentally benign elements.^[20,31–35] Ag₂Te QDs, i.e., QD1 (λ_{1st} = 1100 nm, where λ_{1st} is the distinct first transition peak position in the absorbance spectra), QD2 (λ_{1st} = 1250 nm), QD3 (λ_{1st} = 1400 nm), and QD4 (λ_{1st} = 1730 nm), were directly photopatterned side-by-side using photocrosslinkable ligands (PXLs) containing a 365-nm UV-crosslinkable benzophenone moiety introduced onto the Ag₂Te QD surfaces. The four channels of the multispectral QD photodetector exhibited respective photoresponses across the SWIR range.

2. Results and Discussion

Multispectral SWIR QD photodetectors were fabricated by directly photopatterning four differently sized Ag₂Te QD subpixels side-by-side (Figure 1a), following a cycle that involves QD film deposition, UV-induced selective ligand crosslinking, pattern development to remove the uncrosslinked area of the film, and finally solid-state ligand exchange.^[36–39] Figure 1b depicts a single cycle of the fabrication steps used to fabricate a single sub-

pixel photodetector device. The Ag₂Te QD film was deposited by spin-coating a photocrosslinkable QD ink onto an indium tin oxide (ITO) electrode substrate. The Ag₂Te QD film was photolithographically patterned based on the UV-induced crosslinking reaction of the PXL, followed by the development step, to generate the targeted patterns. To enhance electronic coupling between neighboring QDs, the as-formed crosslinked Ag₂Te QD patterns were subsequently treated with 0.1% (v/v) 1,2-ethanedithiol (EDT) in acetonitrile, replacing the long insulating ligands. These include oleylamine (OLAm), 1-dodecanethiol (DDT) ligands introduced during synthesis, and the PXLs already introduced on the QD surface before the film deposition step. Herein, UV-induced ligand crosslinking was enabled using the PXLs introduced on the Ag₂Te QD surface via a partial ligand exchange step. During this step, a portion of the native ligands attached to the surfaces of the QDs (OLAm or DDT) were replaced by the PXLs, i.e., (4-((10-mercaptodecyl)oxy)phenyl)(phenyl)methanone (O-BP-1-PXL) (Figure 1c).^[37] O-BP-1-PXL contains a benzophenone moiety at one end and a thiol anchor group at the other end. The benzophenone derivative of O-BP-1-PXL imparts the QD films with photocrosslinking capabilities, whereas the thiol anchor group facilitates the binding of the ligands to the surfaces of the QDs upon implementing the ligand exchange step. Notably, adding a silver iodide-oleylamine (AgI-OLAm) precursor to the QD ink was critical for attaining a stable dispersion of QD inks after the partial ligand exchange step.^[33] When 5 wt% of O-BP-1-PXL was added to the ink, 10.7 mol% of the original OLAm and DDT were replaced with O-BP-1-PXL (Figure S1, Supporting Information). The resulting Ag₂Te QDs were cast into solid-state thin films, after which the benzophenone derivatives of O-BP-1-PXL could be photoactivated to form ketyl radicals under 365-nm UV illumination by abstracting a hydrogen atom from the nearby OLAm or DDT ligands (Figure S2, Supporting Information). The resulting radicals form covalent bonds with the ligands of neighboring QDs (Figure 1d). As shown in Figure 1e, different shapes, namely, a square, circle, and line, are formed in the Ag₂Te QD patterns, and micrometer-scale QD patterns are formed with a minimum feature size of 2 μ m (pitch size = 2 μ m). The as-form QD patterns remained intact after solid-state ligand exchange using EDT.^[39]

To investigate the crosslinking efficiency of the Ag₂Te QD film, we measured the film thickness retention ratio (FRR), a ratio of the thicknesses of the crosslinked Ag₂Te QD film before and after exposure to the development solvent (toluene) (Figure S3, Supporting Information). Figure 2a displays a plot of the FRR values of the photocrosslinkable Ag₂Te QD films (with λ_{1st} = 1400 nm) containing 10.7 mol% of O-BP-1-PXL on their surfaces as a function of exposure dose. This indicates that a UV exposure dose of over 5.2 J cm⁻², achievable by irradiating the film for 10 min using a handheld 365-nm UV lamp with a power of 8.72 mW cm⁻², was required to successfully crosslink the Ag₂Te QD film, yielding an FRR of over 0.9.

We subsequently examined the impact of the UV-exposure dose required for direct photopatterning on the performance of the resulting QD photoconductors in terms of responsivity and transient photoresponse. For the comparison, the pristine Ag₂Te QD device and O-BP-1-PXL-exchanged Ag₂Te QD device were prepared by depositing pristine QD and photocrosslinkable QD, respectively, followed by solid-state ligand exchange using EDT.

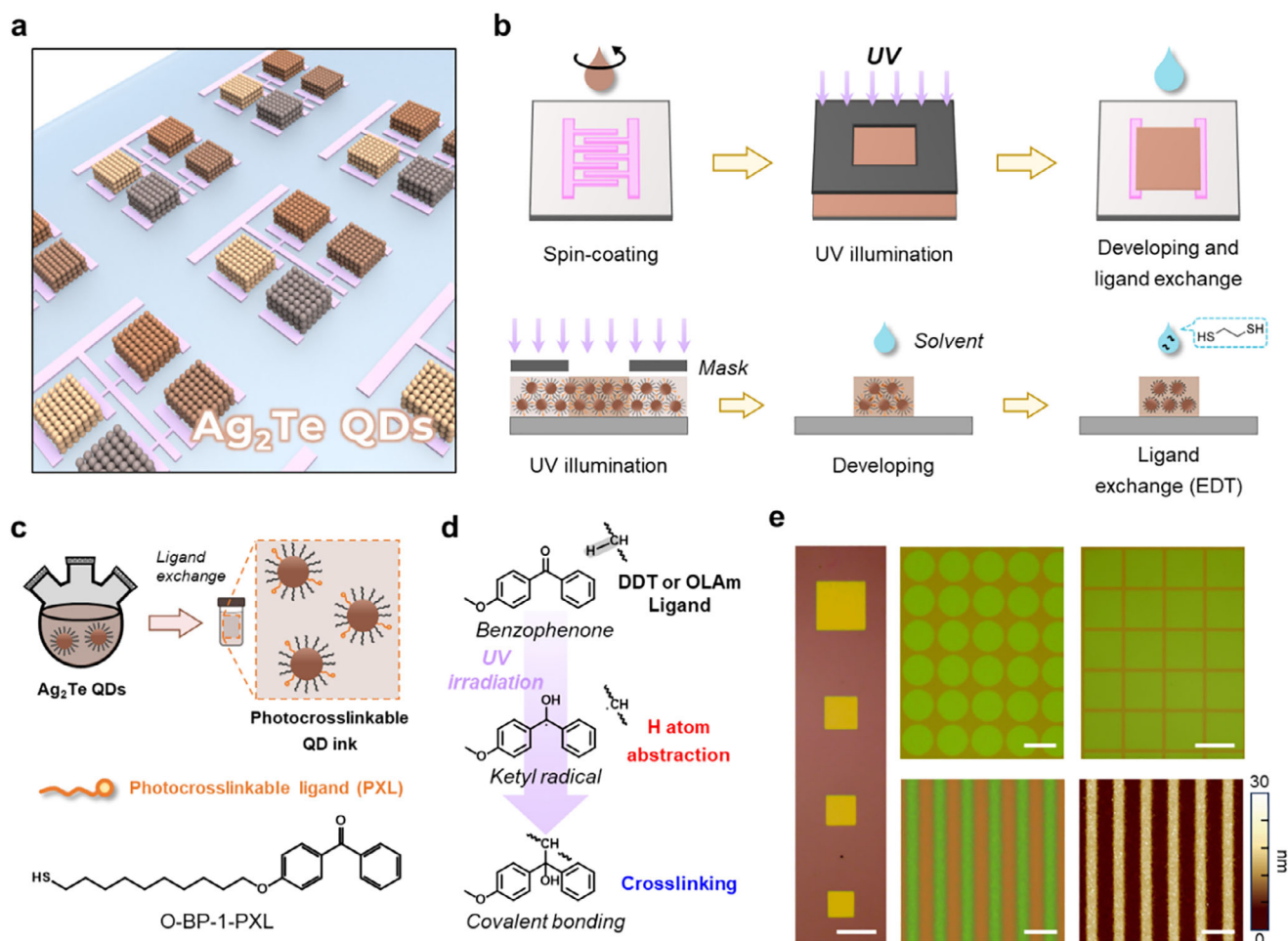


Figure 1. a) Schematic illustration of the multispectral Ag_2Te QD photodetector. b) Schematic illustration showing the fabrication of the patterned Ag_2Te QD photodetector. c) Preparation of the photocrosslinkable Ag_2Te QD ink and chemical structure of O-BP-1-PXL. d) Crosslinking mechanism of the benzophenone moiety present in O-BP-1-PXL with a hydrocarbon group of other ligands (OLAm or DDT ligand). e) Optical microscopy images of the thick square (thickness = 90 nm) (left), circle, square, and line patterns (thickness = 25 nm). The scale bars correspond to 100 (thick square pattern), 20 (circle), 40 (square), and 5 (line) μm . Atomic force microscopy image of the line pattern (scale bar = 5 μm).

Increasing the UV-exposure dose gradually degraded the responsivity of the resulting device (Figure 2b), probably because UV irradiation induced surface oxidation in the outer layer of the Ag_2Te QDs that can act as surface traps. The surface trap defect states can capture photogenerated electrons or holes before they reach the electrodes, leading to increased non-radiative recombination and a faster degradation in photocurrent. The devices based on the O-BP-1-PXL-exchanged Ag_2Te QD films and those based on the pristine Ag_2Te QD film without the partial ligand exchange with O-BP-1-PXL consistently yielded a negative correlation between UV-exposure dose and the responsivity of the patterned device. Interestingly, the former device exhibited relatively higher responsivity at the same UV-exposure dose than the latter. We attribute the superior responsivity of the former device based on the O-BP-1-PXL-exchanged Ag_2Te QD films to the introduction of additional iodide ligands and the PXLs that effectively passivated the surface on the QDs, thereby increasing the robustness against UV illumination. Moreover, the influence of UV-exposure dose on the dynamic responses of the devices was exam-

ined. Figure 2c exhibits the transient photoresponses of the patterned Ag_2Te QD devices fabricated under different UV-exposure doses (0–10.5 J cm^{-2}). As the UV-exposure dose increased, the transient photocurrent response of the patterned Ag_2Te QD devices to 970-nm light signals gradually degraded. The results displayed in Figure 2b,c indicate that although increasing the UV-exposure dose is beneficial for attaining well-defined patterns owing to sufficient crosslinking between the QDs (Figure 2a), the optoelectrical performance of the devices degrades. Therefore, optimizing the UV-exposure dose is important to fabricate multispectral QD detectors with both fine features and good performance via direct photopatterning. In this study, we selected a UV exposure dose of 5.2 J cm^{-2} to fabricate the QD pattern, yielding an FRR of over 0.9 and preserving the responsivity of over 94% (relative to the non-irradiated QD devices) of the crosslinked Ag_2Te QD devices (Figure 2a,b).

Under the optimized photopatterning condition, the IR absorption spectra of the Ag_2Te QD films exhibited no difference after partial ligand exchange with O-BP-1-PXL and

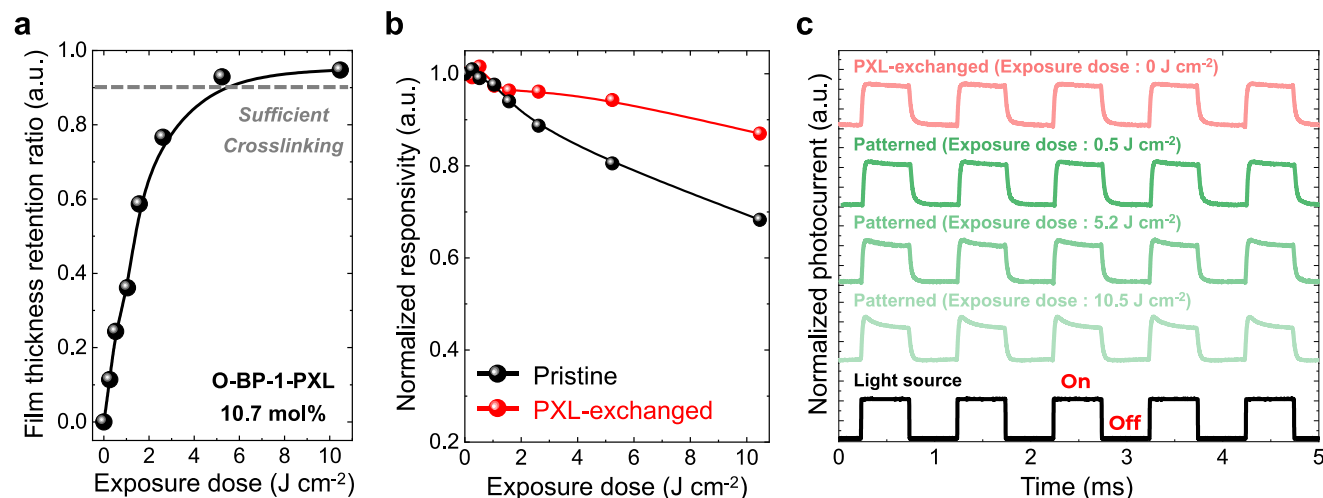


Figure 2. a) FRR of the Ag_2Te QD films crosslinked under various 365-nm UV-exposure doses. b) Normalized responsivity at 1550 nm of the pristine and 5 wt% O-BP-1-PXL-exchanged Ag_2Te QD devices as a function of UV-exposure dose. c) Time-resolved photocurrents of the patterned Ag_2Te QD device under various UV-exposure doses. All devices are solid-state ligand exchanged using EDT.

crosslinking, indicating that light absorption by the Ag_2Te QDs was unaltered during the process (Figure 3a). Likewise, the change in the absorption peaks representing C–H stretching in the Fourier transform infrared (FTIR) spectra of the pristine and O-BP-1-PXL-exchanged Ag_2Te QD films before and after EDT treatment remained almost unaltered. Moreover, 92.6% and 91.3% of the original ligands of the pristine and O-BP-1-PXL-exchanged Ag_2Te QD films were replaced with EDT, respectively (Figure S4, Supporting Information). This similarity suggests that the overall amount of surface-bound aliphatic ligands (containing CH_2 groups) was comparably reduced by EDT treatment in both cases.

Notably, the thickness of the photoconductor channel must be optimized to absorb maximal incident SWIR photons and achieve high responsivity. This can be implemented by the layer-wise deposition of photocrosslinkable Ag_2Te QD ink via cycles of spin coating, patterning, and solid-state ligand exchange with EDT (refer to the Experimental Section). The responsivity spectrum of the patterned Ag_2Te QD photoconductor was enhanced as the number of QD layers deposited increased. A maximum responsivity of 167.3 mA W^{-1} at 1550 nm was obtained when five layers of Ag_2Te QDs were deposited (thickness = 334 nm) (Figure 3b). The photocurrent of the device with five layers of Ag_2Te QDs was $9.4 \times 10^{-6} \text{ A}$ under a bias voltage of 0.2 V at room temperature, whereas the dark current was $6.2 \times 10^{-6} \text{ A}$ (Figure S5, Supporting Information).

We subsequently explored the influence of each step of the photopatterning process on the performance of the device containing the five-layered Ag_2Te QD channels. For this purpose, we fabricated EDT-treated photoconductors based on pristine QD films without introducing O-BP-1-PXL; O-BP-1-PXL-exchanged QD films without UV illumination; and patterned QD films that underwent partial ligand exchange in solution, UV illumination, and development. The performances of these devices were then compared. The patterned device exhibited a responsivity comparable to the other two devices across an IR range of 800–2000 nm (Figure 3c), indicating that the static optoelectrical character-

istics of the QD films were largely unaffected by the introduction of O-BP-1-PXL and the UV-induced crosslinking reactions between the ligands. Consistent trend was shown in an external quantum efficiency (EQE) of these devices (Figure S6, Supporting Information). However, the dynamic response of each device changed according to the transient photocurrent (Figure 3d). Although the O-BP-1-PXL-exchanged QD films exhibited similar dynamic responses to the pristine QD films, the patterned Ag_2Te QD films showed slightly accelerated decay characteristics. We attribute this acceleration to the increases in the trap densities on the surfaces of the QDs owing to the UV-irradiation step, consistent with the results displayed in Figure 2c. High-resolution x-ray photoelectron spectroscopy (XPS) was performed to further investigate the observed degradation of the device performance under UV-irradiation step (Figure S7, Supporting Information). The Te 3d XPS spectrum of the patterned Ag_2Te QD film exhibited additional shoulder peaks at ≈ 575.5 and 585.9 eV after UV-irradiation, indicative of partial oxidation from Te^{2-} to Te^{4+} . This UV-induced oxidation, which likely results in the formation of TeO_2 , on the surface of Ag_2Te QD is known to generate surface traps on the QDs.^[40–41]

In addition to the device performance under UV exposure, the device stability was also assessed in terms of photoresponsivity. First, to explore the air stability of the patterned Ag_2Te QD device, we examined its responsivity at 1550 nm over a two-week period under ambient conditions without encapsulation. Based on the photocurrent measurements, the device retained over 90% of its initial performance after two weeks (Figure S8, Supporting Information). Second, the operational stability of the device was evaluated by subjecting it to continuous illumination with a 1200 nm light source ($\approx 1 \text{ mW cm}^{-2}$). Over an eight-hour period, the photocurrent was observed to increase slightly, indicating that the device is reasonably stable under prolonged exposure to IR light.

Finally, we fabricated multispectral photoconductors via direct photolithography using Ag_2Te QDs with four different sizes and distinct λ_{1st} values in the absorbance spectra as follows: QD1 (diameter = 2.7 nm, $\lambda_{1st} = 1100 \text{ nm}$), QD2 (diameter = 3.5 nm,

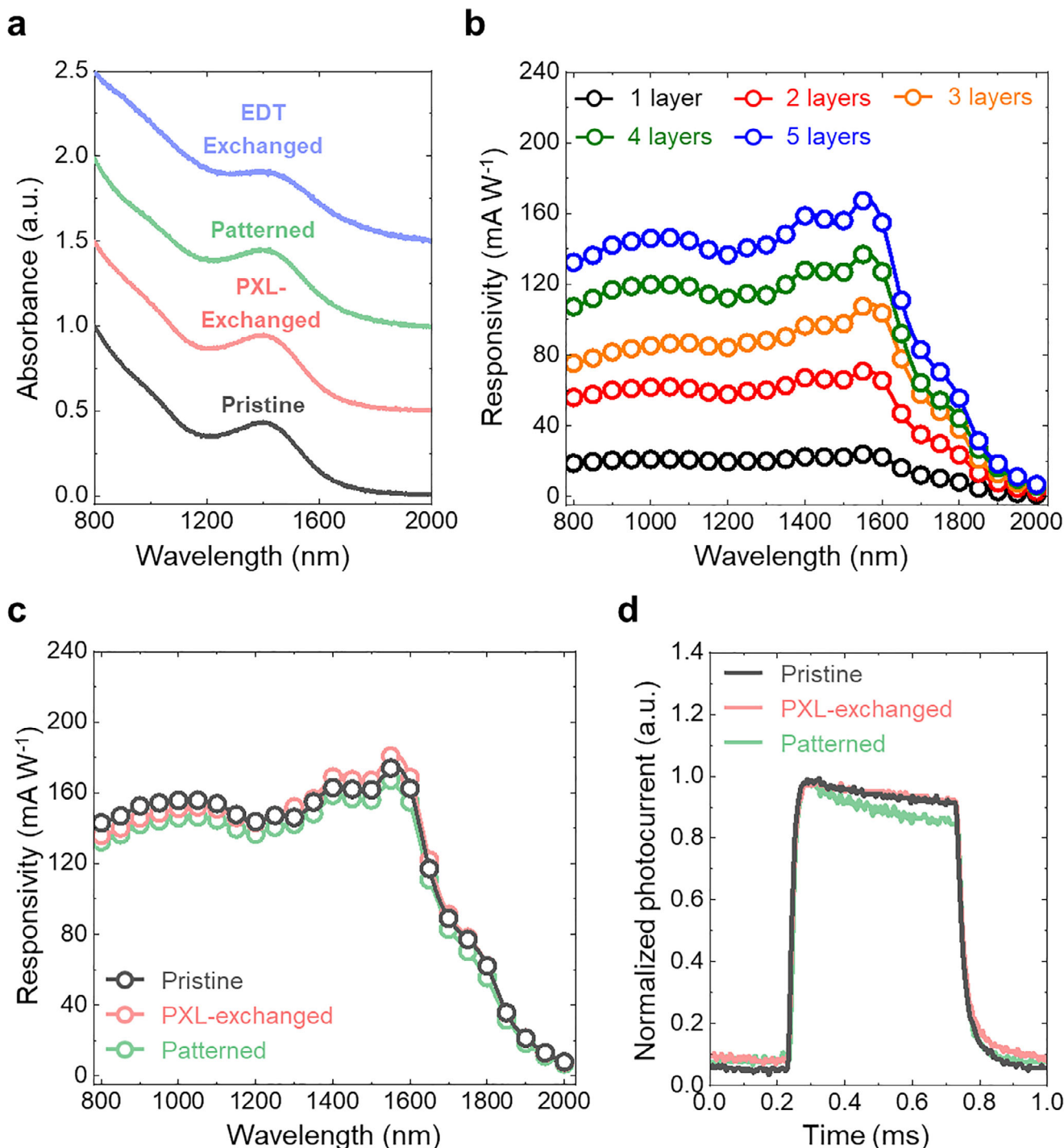


Figure 3. a) Absorption spectra of the pristine, O-BP-1-PXL-exchanged, patterned, and EDT-exchanged Ag₂Te QD films. b) Responsivity spectra of the patterned Ag₂Te QD photoconductors containing various layers under a bias voltage of 0.2 V at a light frequency of 90 Hz. c) Responsivity spectra under a bias voltage of 0.2 V at a light frequency of 90 Hz. d) Time-resolved photocurrents of the pristine, O-BP-1-PXL-exchanged, and patterned Ag₂Te QD devices under a bias voltage of 0.2 V. All devices are solid-state ligand exchanged using EDT.

$\lambda_{1st} = 1250$ nm), QD3 (diameter = 4.1 nm, $\lambda_{1st} = 1400$ nm), and QD4 (diameter = 5.7 nm, $\lambda_{1st} = 1730$ nm) (Figure 4a; Figure S9, Supporting Information). These QDs were synthesized by controlling injection temperature or using a continuous injection method (refer to the Experimental Section for the synthe-

sis details). This set of wavelengths was selected because they encompass the IR range beyond the detection limit of traditional Si-based (≈ 1100 nm) and InGaAs (≈ 1700 nm) photodetectors. These QDs were applied to fabricate the multispectral IR QD photoconductor via four iterations of their photopatterning

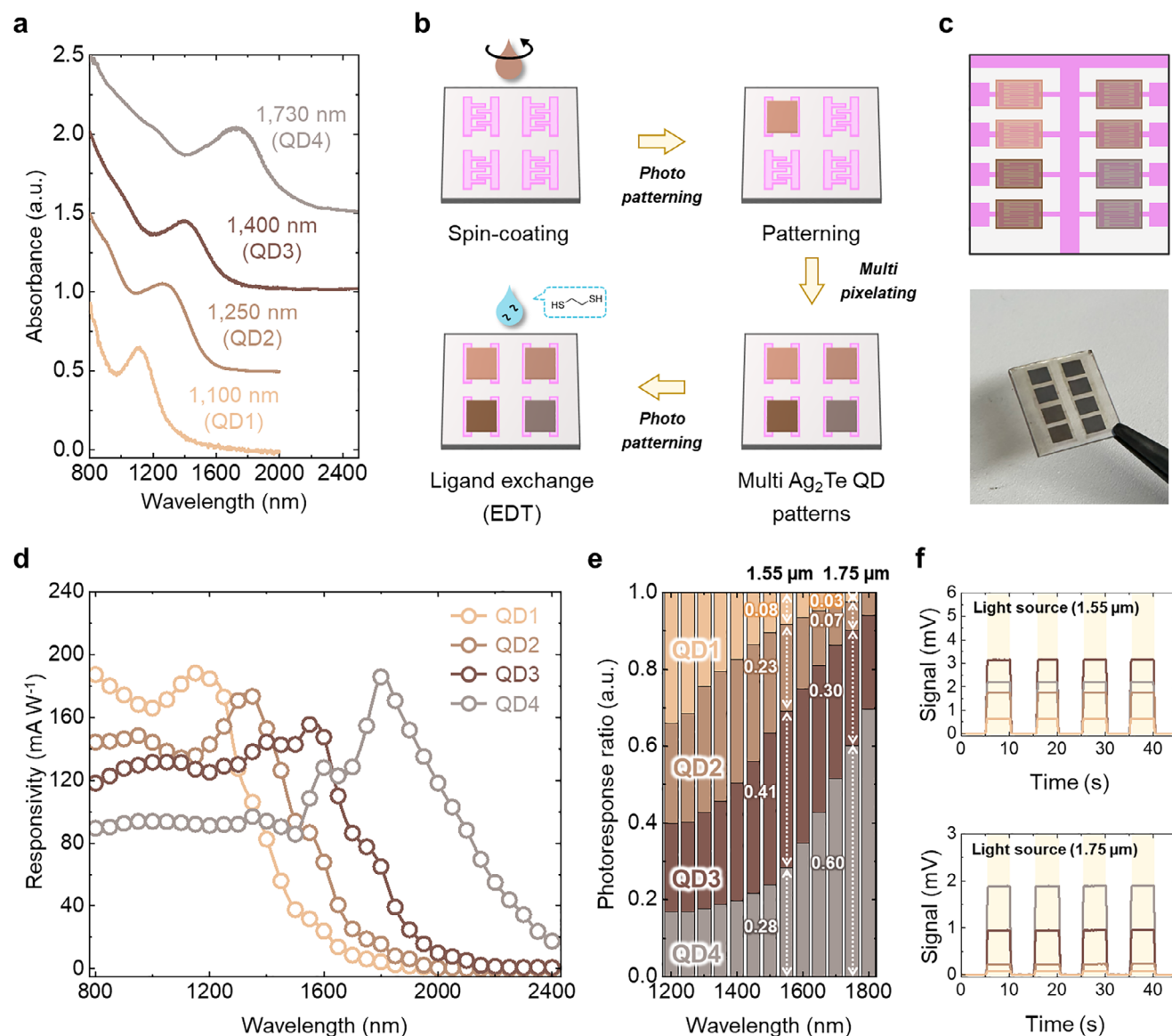


Figure 4. a) Absorption spectra of the four different Ag_2Te QD films (QD1–4). b) Schematic illustration of the fabrication of the patterned multispectral Ag_2Te QD photoconductor using QD1–4. c) Schematic image (top) and photograph (bottom) of the multispectral Ag_2Te QD device. d) Responsivity spectra of the QD1–4 channels on the multispectral device under a bias voltage of 0.2 V at a light frequency of 90 Hz. e) Wavelength decryption chart for spectral range between 1.2 to 1.8 μm derived from the relative signal intensities of QD1–4 channels. f) Measured photoresponse signal upon illumination with 1.55 (left) and 1.75 μm (right) light source.

protocol. Notably, crosslinking the Ag_2Te QD films that developed chemical tolerance against solution processes was beneficial for directly photopatterning the QD film or forming QD patterns side-by-side. If the QDs were not crosslinked, they could easily be removed during the subsequent QD deposition step. Solid-state ligand exchange with EDT was immediately conducted after all the different Ag_2Te QD subpixels were patterned. This process was repeated five times to fabricate thick patterns (>300 nm) to obtain high responsivities (Figure 4b). The schematic of the top-view of the device displayed in Figure 4c reveals the arrangement of four pairs of patterned Ag_2Te QD subpixels on a single substrate. One ITO electrode and a common ITO electrode (center line) served as two contacts via which

a voltage bias can be applied, and the electrical signal can be collected from each Ag_2Te QD pattern. The photograph depicting the bottom view of the actual fabricated device displayed in Figure 4c reveals that all four Ag_2Te QDs were patterned at the desired location. The width and length of the pattern were 3.5 and 2.5 mm, respectively, sufficient to cover the active area of the channel. Figure 4d displays the responsivity spectra of four different Ag_2Te QD channels; the QD1–4 channels yield maximum responsivities of 188.4, 173.5, 155.6, and 185.8 mA W^{-1} and EQE of 20.3%, 15.9%, 12.4%, and 12.8% at 1150, 1350, 1550, and 1800 nm, respectively, under room temperature (Figure S10, Supporting Information). We also measured frequency-dependent noise spectral density (NSD) and specific detectivity (D^*) of the

multispectral QD device (Figure S11, Supporting Information and Experimental Section). All QD1–4 channels show a flicker noise behavior at frequencies below ≈ 2 Hz ($1/f$ noise corner frequency). Above this frequency, the noise is dominated by a white noise floor, which is composed of contributions from generation-recombination, thermal, and shot noise. The noise spectral density of QD1–4 channels exhibit 3.8×10^{-10} , 7.1×10^{-12} , 1.3×10^{-11} , and 6.1×10^{-12} A Hz $^{-0.5}$ at 90 Hz, respectively. We then calculated the D^* of each QD channel. Figure S11c (Supporting Information) presents the room temperature D^* values under a bias voltage of 0.2 V at 90 Hz; QD1–4 channels exhibit maximum D^* of 4.1×10^9 , 2.0×10^9 , 9.9×10^8 , and 2.5×10^9 Jones, respectively.

Using multichannel devices, we demonstrate simple yet informative wavelength identification through a non-spectrometric method. In conventional approaches, identifying the wavelength of incoming light typically requires additional optical components, such as gratings or filters, followed by wavelength-resolved detection or scanning methods.^[42–44] These setups typically involve complex spectrometric measurements and often bulky instrumentation.

In contrast, our 4-channel device enables wavelength recognition based on a simple signal readout, without any additional optics. As shown in Figure 4d, each QD channel (QD1–QD4) exhibits a unique spectral responsivity profile. In the longer wavelength region above 2100 nm, for instance, only QD4 shows a significant response. However, in the wavelength region between 1200 and 1800 nm, all four channels respond to varying degrees, each with distinct relative responsivity levels depending on the wavelength. We summarize this result in a wavelength decryption chart (Figure 4e), which maps the normalized signal ratios from the four channels to their corresponding input wavelengths. When SWIR between 1200 and 1800 nm within the linear dynamic range is incident on the device, it generates a specific combination of photoresponses across the four channels. By comparing this signal pattern to the decryption chart, one can deduce the input wavelength.

As a demonstration, we show two representative examples in Figure 4f. In one case, the QD1:2:3:4 photovoltage ratios were measured as 0.64 mV:1.75 mV:3.14 mV:2.19 mV, corresponding to 8:23:41:28, and in another case as 0.08 mV:0.23 mV:0.95 mV:1.89 mV, corresponding to 3:7:30:60. By referencing these ratios to the decryption chart in Figure 4e (dashed white lines in the figure), the input wavelengths were determined to be ≈ 1550 and 1750 nm, respectively. Notably, no other wavelengths within the 1200–1800 nm range produce the same combination of photovoltage ratios. Potential ambiguity in identifying the incident wavelength—arising from different wavelengths generating similar photovoltage ratios—can be mitigated by incorporating additional channels with distinct bandgaps into the multispectral device. This method of wavelength recognition is a unique and effective capability enabled by the multispectral configuration, which is not achievable with a single-channel QD detector. We comment that the accuracy of this method depends on the resolution and diversity of the decryption chart. The horizontal resolution (x -axis) is determined by the wavelength step size used in calibrating the spectral responsivity (50 nm in our case), while the vertical resolution (y -axis) depends on the degree of variation in the responsivity ratios among the channels. Increasing the number of spectrally distinct channels would further enhance

the richness of signal combinations, thereby improving the precision of wavelength identification.

3. Conclusion

We demonstrated the direct photopatterning of heavy-metal-free Ag₂Te QDs using photocrosslinkable ligands to fabricate a multispectral SWIR QD photodetector containing Ag₂Te QDs with four different sizes. Integrating the four QDs with different spectral responses via direct photopatterning enables multispectral IR photodetection, which could be exploited for demonstrating simple wavelength identification through a non-spectrometric method. The fabricated device indicates that this method is simple and feasible for realizing multispectral QD photodetectors that simultaneously respond to various IR sources. This approach is general and can be easily adapted to other QD systems operating through various energy ranges across the entire electromagnetic spectrum. Furthermore, the direct photopatterning method can impart high-resolution pixelation of the IR QDs integrated onto a CMOS ROIC, allowing an IR imager with excellent spatial resolution to be realized and paving the path for multispectral and hyperspectral IR imaging.

4. Experimental Section

Materials: AgI (99.9%) was purchased from Bean Town Chemical. TeO₂ (99.9995%), OLAm (technical grade), 1-octadecene (ODE, 99%), DDT ($\geq 98\%$), K₂CO₃ ($>99\%$), and NaSH ($\geq 60\%$) were purchased from Sigma-Aldrich. Hydroquinone ($>99\%$), 1,10-dibromodecane ($>95\%$), and tetrabutylammonium bromide (TBAB, $>98\%$) were purchased from Tokyo Chemical Industry. OLAm was used after degassing at 110 °C under vacuum. All the other chemicals were used without further purification.

Synthesis of the Ag₂Te QDs: The Ag₂Te QDs were synthesized using previously reported methods with minor modifications.^[20] A 1 M AgI-OLAm precursor solution was prepared by mixing 10 mmol AgI and 10 mL OLAm at 50 °C in an inert atmosphere. The Te precursor, i.e., 2.5 mmol TeO₂ and 10 mL DDT, was loaded into a three-neck flask and degassed at 80 °C for 30 min. The flask was then filled with nitrogen and heated to 100 °C to fully dissolve the TeO₂ into the DDT. The final synthesis of the Ag₂Te QDs involved loading 30 mL of OLAm and 15 mL of ODE in the reaction flask and degassing at 110 °C for 4 h to remove oxygen and moisture. The reaction flask was then filled with nitrogen and heated to the desired temperature to obtain the differently sized QDs. Once the desired temperature was reached, 3 mL of the AgI-OLAm precursor was injected into the reaction flask. After the temperature reached the desired point, 5 mL of the TeO₂-DDT precursor was rapidly injected into the flask. After 15 min of reaction, the heat was turned off, and the reaction flask was cooled naturally. The as-synthesized Ag₂Te QDs were purified using toluene and methanol according to the previous method.^[20] The final solid Ag₂Te QD precipitates were dispersed in chloroform. To synthesize the Ag₂Te QDs with a first excitonic peak of 1730 nm, the AgI-OLAm and TeO₂-DDT precursors were injected at 140 °C and reacted for 15 min, similar to the synthesis of the QDs with different sizes. Subsequently, 1.0 mL of AgI-OLAm and 1.8 mL of the TeO₂-DDT precursor were continuously injected using a syringe pump with an injection speed of 0.05 mL min⁻¹. After three cycles of continuous injection, Ag₂Te QDs with an excitonic peak of 1730 nm were obtained.

To prepare the photocrosslinkable Ag₂Te QD ink, 5 wt% of O-BP-1-PXL in chloroform was added to the Ag₂Te QD solution containing the same number of moles of the AgI-OLAm precursor. The mixture was stirred for 1 h and purified via centrifugation to remove any precipitates. The supernatant was centrifuged by adding methanol and redispersed in chloroform.

Synthesis of O-BP-1-PXL: O-BP-1-PXL was synthesized by following a previous report.^[37] Hydroquinone (15 g), 1,10-dibromodecane (34.1 g), and K₂CO₃ (16 g) were loaded into a flask with 400 mL of anhydrous acetone. The flask was heated to 60 °C, and the temperature was maintained for 9 h before the mixture was cooled to room temperature. The mixture was filtered and washed with deionized water, and the reactant was extracted using methylene chloride. The organic layer was dried over MgSO₄ and concentrated under reduced pressure. The unreacted reagent was removed by distilling the residue, which was purified via column chromatography (4:1 *n*-hexane:ethyl acetate). The collected white solid was loaded into a flask with NaSH (10.7 g), TBAB (4.61 g), and 160 mL of anhydrous dimethylformamide. The flask was heated to 120 °C and maintained for 6 h. After quenching the reaction with NH₄Cl, the precipitate was purified via column chromatography (4:1 *n*-hexane:ethyl acetate) to obtain O-BP-1-PXL as an off-white solid.

Device Preparation: All the device fabrication steps were executed in ambient air. The ITO electrode substrates were cleaned via ultrasonication in deionized water, acetone, and isopropanol for 15 min each. After drying with nitrogen, the substrates were UV/ozone treated for 30 min. To obtain each layer of the patterned Ag₂Te QD film, the photocrosslinkable Ag₂Te QD ink solution with a concentration of 100 mg mL⁻¹ was spin-coated onto an ITO substrate (1000 rpm for 60 s). The resulting Ag₂Te QD film was crosslinked under UV illumination (365-nm, 8.72 mW cm⁻²). Subsequently, the crosslinked Ag₂Te QD film was dipped into toluene for 3 s. After fully drying the film, 0.1% (v/v) EDT in acetonitrile was applied to the crosslinked Ag₂Te QD film for 30 s. Next, the substrate was rinsed by subsequently spin-coating acetonitrile and toluene at 2000 rpm for 30 s. To obtain the multilayers, the previous steps were repeated. Finally, the film was annealed at 110 °C for 10 min and dried under vacuum for 30 min to remove any residual solvent.

To fabricate the multispectral Ag₂Te QD photoconductor, photocrosslinkable QD1–4 inks were prepared with a concentration of 100 mg mL⁻¹. Each photocrosslinkable QD ink was spin-coated onto a pre-cleaned ITO substrate and patterned using the same procedure as that described above. Once QD1–4 was patterned, solid-state ligand exchange with 0.1% (v/v) EDT in acetonitrile was implemented collectively before depositing the next QD layer to ensure the equal degree of ligand exchange with EDT. After depositing five layers of QD1–4, the device was annealed at 110 °C for 10 min and subsequently dried under vacuum for 30 min to remove any residual solvent.

Absorbance and FTIR: The absorption spectra of the pristine, O-BP-1-PXL, and patterned Ag₂Te QDs were measured using a CARY 5000 UV-visible-near IR (UV–vis–NIR) spectrometer using a clean CaF₂ substrate. To explore the solid-state ligand exchange with EDT, a Nicolet 6700 FTIR spectrometer was used, and the Ag₂Te QD samples were prepared similarly to those prepared for UV–vis–NIR spectroscopy.

Responsivity and EQE: The responsivity of the Ag₂Te QD device was characterized using a custom-fabricated visible-to-SWIR photoconductivity setup. Broadband light (300–3800 nm) from an incoherent 250 W Oriel Newport Light source equipped with a halogen bulb was subsequently filtered and modulated at 90 Hz. A ¼ m monochromator with 39-nm resolution was employed for spectral selection, complemented by automatically adjusted high-pass filters. The monochromatic output was focused on the sample, housed in a light-tight enclosure. Incident power was quantified using a calibrated Ge detector (818-ST2-IR). The photocurrents were amplified and converted to voltage via a current preamplifier (Stanford Research Systems SR570) and lock-in amplifier configuration (SR810). Data were acquired and analyzed using custom LabVIEW software, enabling the spectral photoresponse to be comprehensively evaluated. The spectral responsivity and EQE were calculated using the following equations: $R = I_{ph}/P = EQE \times \lambda / 1240$ where R is the responsivity (A W⁻¹), I_{ph} is the generated photocurrent (A), P is the incident optical power (W), EQE is an external quantum efficiency (%), and λ is the wavelength (nm).

Noise Spectral Density (NSD) and Specific Detectivity (D*): Noise spectral density was measured using a femto DLCPA-200 preamplifier and an SR770 spectrum analyzer. The device is mounted in a black-out ST500 cryostat with short triaxial cables fed into the preamplifier. 200 linear averages were used for each scan. All instruments are grounded to a common

ground using copper wiring. The spectral detectivity was calculated using the following equation: $D^* = R \times \sqrt{A \times \Delta f} / i_n$ where A is the device area (0.00665 cm²), Δf is the bandwidth (1 Hz), and R and i_n represent the responsivity (A W⁻¹) and the noise spectral density (A Hz^{-0.5}).

Current–Voltage Characteristics: A Keithley 2400 source meter was used to measure the voltage-dependent dark and photocurrent (I – V) characteristics under a 970-nm IR source (M970L4).

Time-Resolved Photocurrent: An oscilloscope (TDS 1002) with a 970-nm light-emitting diode source was used to measure the time-resolved photocurrents of the samples. The signal was amplified using a preamplifier (SR570).

Patterning and Atomic Force Microscopy: High-resolution patterns of the Ag₂Te QDs were fabricated using a Karl Süss MA-6 Mask Aligner. The FRR, surface morphologies, and height profiles of the patterned Ag₂Te QD films were obtained using a Park NX10 atomic force microscopy system (Park Systems) under ambient conditions in the non-contact mode.

¹H-Nuclear Magnetic Resonance (¹H-NMR): The ¹H-NMR spectra of the samples were measured using a Bruker Avance III HD spectrometer at 500 MHz with deuterated chloroform as the solvent.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

A.S. and M.S.K. conceived the concept and designed the experiment. S.Y.P. and E. H. conducted the Ag₂Te QD synthesis. S.Y.P. and S.J.P. conducted the electrical measurement and the absorption spectroscopy measurement. S.Y.P., Y.H.J., Z.L., H.J., H.Z., Y. A., and J.H.K. performed the material characterization. S.Y.P., S.L., and Y.H.J. conducted Ag₂Te QD patterning and topological analysis. S.Y.P. wrote the manuscript under supervision of A.S. and M.S.K.

Data Availability Statement

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

Keywords

crosslinking, infrared, multispectral, patterning, photodetector, quantum dot

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- [1] J. Guo, S. Gu, L. Lin, Y. Liu, J. Cai, H. Cai, Y. Tian, Y. Zhang, Q. Zhang, Z. Liu, Y. Zhang, X. Zhang, Y. Lin, W. Huang, L. Gu, J. Zhang, *Nat. Commun.* **2024**, *15*, 5193.
- [2] Z. Wang, Y. Gu, X. Li, Y. Liu, F. Liu, W. Wu, *Adv. Opt. Mater.* **2023**, *11*, 2300970.
- [3] U. Adomeit, J. Krieg, *SPIE* **2015**, 9641, 16.
- [4] D. J. Naczynski, M. C. Tan, M. Zevon, B. Wall, J. Kohl, A. Kulesa, S. Chen, C. M. Roth, R. E. Riman, P. V. Moghe, *Nat. Commun.* **2013**, *4*, 2199.
- [5] E. D. Cosco, A. L. Spearman, S. Ramakrishnan, J. G. P. Lingg, M. Saccomano, M. Pengshung, B. A. Arús, K. C. Y. Wong, S. Glasl, V. Ntziachristos, M. Warmer, R. R. McLaughlin, O. T. Bruns, E. M. Sletten, *Nat. Chem.* **2020**, *12*, 1123.
- [6] Z. Liu, H. Ukida, P. Ramuhalli, K. Niel, *Advances in Computer Vision and Pattern Recognition*, Springer Nature, Berlin, Germany, **2015**.
- [7] P. LeVan, U. Sakoglu, *SPIE* **2020**, 11503, 38.
- [8] Z. Wang, B. Li, L. Li, *IOP Conf. Series: Earth Environm. Sci.* **2019**, 237, 032087.
- [9] Z. Li, S. Trendafilov, F. Zhang, M. S. Allen, J. W. Allen, S. U. Dev, W. Pan, Y. Yu, Q. Gao, X. Yuan, I. Yang, Y. Zhu, A. Bhat, S. X. Peng, W. Lei, H. H. Tan, C. Jagadish, L. Fu, *Nano Lett.* **2021**, *21*, 7388.
- [10] Y. Wang, K. Dong, Y. Song, M. Piao, B. Zhang, L. Zhang, G. Yan, Z. Liang, T. Liu, X. Li, *Opt. Commun.* **2025**, 575, 131282.
- [11] "HAMAMATSU, Si/InGaAs two-color detector, https://www.hamamatsu.com/content/dam/hamamatsu-photonics/sites/documents/99_SALES_LIBRARY/ssd/k1713-003_kird1132e.pdf,"
- [12] "HAMAMATSU, Si/InGaAs two-color detector, https://www.hamamatsu.com/content/dam/hamamatsu-photonics/sites/documents/99_SALES_LIBRARY/ssd/k3413-05_etc_kird1041e.pdf,"
- [13] S. Woo, E. Yeon, H.-Y. Ryu, J.-H. Han, H. W. Jang, D. Jung, W. J. Choi, *Adv. Funct. Mater.* **2025**, 35, 2419329.
- [14] E. R. Blazejewski, J. M. Arias, G. M. Williams, W. McLevege, M. Zandian, J. Pasko, *J. Vacuum Sci. Technol. B* **1992**, *10*, 1626.
- [15] G. Ariyawansa, J. Duran, M. Grupen, J. Scheihing, T. Nelson, M. Eismann, *SPIE* **2012**, 8353, 154.
- [16] M. Razeghi, A. Dehzangi, J. Li, *Results Opt.* **2021**, *2*, 100054.
- [17] P. Mitra, F. C. Case, J. H. McCurdy, S. A. Zaidel, L. T. Claiborne, *Appl. Phys. Lett.* **2003**, *82*, 3185.
- [18] L. Meng, Ji Zhou, S. Liu, L. Ding, J. Zhang, S. Wang, T. Lei, *Int. J. Appl. Earth Observat. Geoinform.* **2021**, *102*, 102403.
- [19] A. Rogalski, *Opto-Electron. Rev.* **2012**, *20*, 279.
- [20] Y. Wang, L. Peng, J. Schreier, Yu Bi, A. Black, A. Malla, S. Goossens, G. Konstantatos, *Nat. Photonics* **2024**, *18*, 236.
- [21] S. Keuleyan, E. Lhuillier, V. Brajuskovic, P. Guyot-Sionnest, *Nat. Photonics* **2011**, *5*, 489.
- [22] C. Livache, B. Martinez, N. Goubet, C. Gréboval, J. Qu, A. Chu, S. Royer, S. Ithurria, M. G. Silly, B. Dubertret, E. Lhuillier, *Nat. Commun.* **2019**, *10*, 2125.
- [23] M.-J. Si, D. Kim, S. Jeong, M. Yang, J. Kim, S.-Y. Lee, I.-S. Lee, J. Jeong, B.-C. Kim, T. Han, B. Kim, Y. Ahn, S. Jee, Y. Jung, S.-W. Baek, *Korean J. Chem. Eng.* **2024**, *41*, 3561.
- [24] S. Ahn, J. Y. Shang, S. K. Patel, E. Alvarado, A. Ndao, O. Vazquez-Mena, *Adv. Funct. Mater.* **2024**, *34*, 2409523.
- [25] X. Tang, M. M. Ackerman, M. Chen, P. Guyot-Sionnest, *Nat. Photonics* **2019**, *13*, 277.
- [26] G. Mu, Y. Tan, C. Bi, Y. Liu, Q. Hao, X. Tang, *Nat. Photonics* **2024**, *18*, 1147.
- [27] S. Zhang, C. Bi, Y. Tan, Y. Luo, Y. Liu, J. Cao, M. Chen, Q. Hao, X. Tang, *ACS Nano* **2022**, *16*, 18822.
- [28] J. Kim, S.-M. Kwon, Y. K. Kang, Y.-H. Kim, M.-J. Lee, K. Han, A. Facchetti, M.-G. Kim, S. K. Park, *Sci. Adv.* **2019**, *5*, aax8801.
- [29] A. Ali, S. Oh, W. Kim, S. J. Oh, *Korean J. Chem. Eng.* **2024**, *41*, 3545.
- [30] Y. Sung, J. Chang, S. Choi, S. Jeong, *Korean J. Chem. Eng.* **2024**, *41*, 3317.
- [31] Z.-Y. Liu, A.-A. Liu, H. Fu, Q.-Y. Cheng, M.-Y. Zhang, M.-M. Pan, L.-P. Liu, M.-Y. Luo, B. Tang, W. Zhao, J. Kong, X. Shao, D.-W. Pang, *J. Am. Chem. Soc.* **2021**, *143*, 12867.
- [32] G. Kim, D. Choi, S. Y. Eom, E. D. Jung, J. H. Lee, B. Rehl, S. Y. Kim, S. Hoogland, E. H. Sargent, K. S. Jeong, *ACS Mater. Lett.* **2024**, *6*, 4988.
- [33] Z.-Y. Liu, W. Zhao, L.-M. Chen, Y.-Y. Chen, Z.-G. Wang, An-An Liu, D.-W. Pang, *Nano Res.* **2024**, *17*, 10585.
- [34] Y. Wang, H. Wu, C. Rodà, L. Peng, N. Taghipour, M. Dosil, G. Konstantatos, *Adv. Mater.* **2025**, 27, 2500977.
- [35] D. Yuan, F. Cao, Z. Han, M. Liu, X. Deng, Li Zhang, X. Liu, S. Cao, X. Liu, Z. Yang, Z. Deng, Y. Li, O. Voznyy, Q. Fan, B. Sun, W. Huang, *Nano Energy* **2025**, *140*, 111026.
- [36] J. Yang, M. Lee, S. Y. Park, M. Park, J. Kim, N. Sitapure, D. Hahm, S. Rhee, D. Lee, H. Jo, Y. H. Jo, J. Lim, T. J. Shin, D. C. Lee, K. Kwak, J. S. Kwon, B. Kim, W. K. Bae, M. S. Kang, *Adv. Mater.* **2022**, *34*, 2205504.
- [37] D. Hahm, J. Lim, H. Kim, J.-W. Shin, S. Hwang, S. Rhee, J. H. Chang, J. Yang, C. H. Lim, H. Jo, B. Choi, N. S. Cho, Y.-S. Park, D. C. Lee, E. Hwang, S. Chung, C.-M. Kang, M. S. Kang, W. K. Bae, *Nat. Nanotechnol.* **2022**, *17*, 952.
- [38] S. Y. Park, S. Lee, J. Yang, M. S. Kang, *Adv. Mater.* **2023**, *35*, 2300546.
- [39] J. Il Yoon, H. Kim, M. Kim, H. Cho, Y. A. Kwon, M. Choi, S. Park, T. Kim, S. Lee, H. Jo, B. Kim, J. Ho Cho, J.-S. Park, S. Jeong, M. S. Kang, *Sci. Adv.* **2023**, *9*, adj8276.
- [40] H. Baek, S. Kang, J. Heo, S. Choi, R. Kim, K. Kim, N. Ahn, Y.-G. Yoon, T. Lee, J. B. Chang, K. S. Lee, Y.-G. Park, J. Park, *Nat. Commun.* **2024**, *15*, 1671.
- [41] H. Moon, C. Lee, W. Lee, J. Kim, H. Chae, *Adv. Mater.* **2019**, *31*, 1804294.
- [42] T. Xu, Y.-K. Wu, X. Luo, L. J. Guo, *Nat. Commun.* **2010**, *1*, 59.
- [43] S.-W. Wang, C. Xia, X. Chen, W. Lu, M. Li, H. Wang, W. Zheng, T. Zhang, *Opt. Lett.* **2007**, *32*, 632.
- [44] A. Nitkowski, L. Chen, M. Lipson, *Opt. Express* **2008**, *16*, 11930.