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All inorganic mixed halide perovskite nanocrystal-graphene hybrid photodetector: from ultrahigh gain to photostability

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Abstract

Hybrid graphene-perovskite photodetectors embrace the excellent photoabsorption properties of perovskites and high carrier mobility of graphene in a single device. Here, we demonstrate the integration of halide ion exchanged CsPbBr_xI_{3-x} nanocrystals as photoabsorber and graphene as transport layer. The nanocrystals conform to a cubic lattice structure and exhibit an optical band gap of 1.93 eV. The hybrid device attained a maximum responsivity of 1.13×10^4 A/W and specific detectivity of 1.17×10^{11} Jones in low light intensity ($\sim 80 \mu\text{W}/\text{cm}^2$). Specifically, an ultrahigh photoconductive gain of 9.32×10^{10} is attained due to fast hole transit time in the graphene transistor and long recombination lifetime in the perovskite NCs simultaneously. The phototransistor also shows good stability and can maintain $\sim 95\%$ of the photocurrent under continuous illumination over 5 hrs and $\sim 82\%$ under periodic illumination over 37 hrs. Our results also revealed that the common issue of ion separation and segregated halide domains in mixed halide perovskite NCs do not occur under low light intensities. The intensive degradation of CsPbBr_xI_{3-x} nanocrystals is only observed under stronger light excitation ($\geq 55 \text{ mW}/\text{cm}^2$), reflecting as emission shifts. Our work establishes the use of fully inorganic perovskite NCs as highly stable photodetectors with high responsivity and low power light detection.

Keywords: *photodetector, perovskite nanocrystals, graphene, stability, high-responsivity*

Introduction

Semiconductor nanocrystals (NCs) or quantum dots have been of interest in optoelectronic devices due to their high photoluminescence quantum yield, size dependent bandgap and narrow emission wavelength, as well as good ambient stability through ligand stabilization¹⁻³. Recently, perovskite nanocrystals (NCs), represented by the simple building block, ABX_3 , where A is an organic or inorganic cation (eg. ammonium, cesium), B is the metal cation, and X is a halide anion, have demonstrated excellent photo-absorption due to low bandgap with large carrier diffusion lengths, leading to high photocurrent³⁻⁵ and exhibited excellent narrow emission quantum yield reaching over 90%¹. Furthermore, the perovskite nanocrystals can be easily processed into high quality films, allowing them to be potential candidate materials for various optoelectronic devices such as photovoltaics⁶⁻⁸, light-emitting diodes^{9,10}, photodetectors^{1,11} and lasers^{12,13}. In particular, photodetectors have attracted significant interest due to their broad applications in imaging, security and communications¹⁴⁻¹⁸.

To date, several groups have been successful in producing functional photodetectors using perovskite NCs^{11,19-22}. However, due to the poor charge transport properties and therefore low responsivity of these devices, many studies have integrated perovskite NCs with high mobility 2D materials, including graphene and MoS_2 , where the perovskite NCs act as the sensitizing element and the 2D materials as the transport layer^{23,24}, in a phototransistor configuration to increase responsivity. Photodetection at extremely low incident light intensities can also be achieved through the formation of a sensitizer-conductor hybrid structure which allows effective charge separation induced by the intrinsic band bending at the interface and injection of only one type of charge carrier^{4,23,25-28}. A high gain can also be achieved through the photogating effect, which cause recirculation of the carriers in the channel of a phototransistor multiple times before recombining with the opposite charges. However, phototransistors based on perovskite NCs have shown relatively lower gains ($\sim 10^6$)²⁹ as compared to devices with perovskite bulk films or other functional materials ($\sim 10^7$)³⁰⁻³².

Among the various perovskite compositions, $CH_3NH_3PbX_3$ and $CsPbBr_3$ are well-studied for photodetecting applications³³⁻³⁶. The all-inorganic perovskites are gaining increasing attention since organic-inorganic hybrid perovskites typically suffer from stability issues in humid and ambient conditions³⁷. Furthermore, the red emitting $CsPbI_3$ NCs have been showed to have a relatively longer radiative lifetime than the green and blue emitting NCs which could be beneficial in producing large photocurrents³⁸. But there are seldom reports for

the case of CsPbI₃ NCs based photodetectors¹ as compared to CsPbBr₃ NCs. One of the main reasons might be the inherent phase instability in CsPbI₃, transforming from cubic to undesired orthorhombic phase^{11,16,39}. Surface ligands of semiconductor nanocrystals play important roles in maintaining its stability and controlling the physical/chemical properties⁴⁰. The surface ligands control the nucleation, growth dynamics as well as determine the final phase^{38,41}. It has been suggested that the conventionally used ammonium based ligands form weak acid-base interactions with I⁻ in the CsPbI₃ NCs, causing easy loss of the ligands especially during the purification process and lead to fast agglomeration and undesired phase transformation^{6,7,42}. While the community is searching for new NC synthesis with different capping ligands, another straightforward approach at forming stable and higher responsivity is to utilise mixed halide CsPb(Br/I)₃ and purposefully tune the absorption of the perovskite NCs towards longer wavelength through the control of halide content (I/Br ratio). In particular, the use of anion exchange reaction allows fine control over the nanocrystal composition⁴³.

In this work, we report for the first time, the application of CsPbBr_xI_{3-x} NCs with higher iodide to bromide ratio as photo-absorption layer and graphene as transport layer. Unlike previous report using CsPbBr_xI_{3-x} NCs with lower iodide to bromide ratio (emission peak is at 604nm),²⁴ the PL emission and absorption edge of our NCs shifted to longer wavelengths (650 nm). Stable, high-performance photoconductive detector with good photoresponsivity of 1.13×10^4 A/W and specific detectivity of 1.17×10^{11} Jones in low light intensity ($\sim 80 \mu\text{W}/\text{cm}^2$) have been achieved in the CsPbBr_xI_{3-x} NCs-graphene hybrid structures. An ultra-high photoconductive gain of 9.32×10^{10} is also attained due to the ultrafast transit time of holes in graphene channel and the long recombination lifetime of the perovskite NCs simultaneously. These unique properties are verified via Raman spectroscopy and transient photoluminescence measurements. The phototransistor also demonstrated good stability with photocurrent retention of $\sim 95\%$ under continuous illumination over 5 hrs and $\sim 82\%$ under periodic illumination over 37 hrs. Furthermore, our results clarified the effect of light intensities on the stability of the mixed halide NCs and revealed that the common issue of ion separation and segregated halide domains in mixed halide perovskite NCs does not occur under low light intensities. This confirms the potential of the mixed halide NCs for photodetection applications and in particular, towards weak light sensing. Ours results therefore reveal a promising approach for integrating perovskite NCs in photonic devices such as photodetectors, and low power indoor monitoring for internet of things.

Experimental section

Synthesis of $\text{CsPbBr}_x\text{I}_{3-x}$ Nanocrystals (NCs): $\text{CsPbBr}_x\text{I}_{3-x}$ NCs were prepared from CsPbBr_3 NCs followed by anion exchange. Cesium carbonate (0.203 g, 99%) and oleic acid (1.2 mL, 90%) were added to a two neck round bottom flask with glass stopper, followed by vacuum heating at 120° C for one hour. As the cesium carbonate dissolves completely to form cesium oleate, the temperature was further raised to 150° C with purging of N_2 gas. The reaction was then stopped after 30 minutes. For synthesising CsPbBr_3 NCs, PbBr_2 (0.138 g, 99.99%), octadecene (10 mL, 90%), oleic acid (1 mL) and oleylamine (1 mL) were added to a two neck round bottom flask with glass stopper. The PbBr_2 was allowed to dissolve completely at 120 °C under vacuum for about 1 hour. After that, the temperature was increased to 180 °C under N_2 and the cesium oleate solution (800 uL, synthesized in the above steps, pre-heated to 120 °C) was injected. Immediately after the injection of cesium oleate, the reaction mixture was quenched using an ice-water bath. The NCs in octadecene were isolated by centrifugation at 8000 rpm for 10 minutes. The precipitate collected was dispersed in toluene and washed with acetonitrile to wash away the excess ligands. The final precipitate was re-dispersed in 5 mL of toluene.

The precursor for iodide source PbI_2 (0.1 mM, 99.99%) was mixed with octadecene (5 mL), oleic acid (200 uL) and oleylamine (200 uL). After complete solubilisation of PbI_2 at 120 °C under vacuum for about 1 hour, the reaction was stopped with a final purging of N_2 gas and allowed to cool to room temperature. To this reaction mixture, 5 mL of CsPbBr_3 NCs in toluene was added and stirred for 5-10 minutes at room temperature. The mixed halide NCs were first collected by centrifugation and then finally redispersed in toluene, which was used as the active layer for device fabrication.

Device fabrication and characterization: Atomic-layer graphene flakes were mechanically exfoliated from a crystal of highly oriented pyrolytic graphite (HOPG) using adhesive 3M-tape and transferred on a silicon wafer with a 285 nm thermally grown SiO_2 layer. The graphene-based field effect transistors (FETs) were fabricated with the heavily doped silicon substrate as a bottom gate electrode and gold as the source drain electrodes by standard photolithography and e-beam evaporation. Finally, the perovskite NCs was spin coated onto the graphene device at 2000 RPM for 40 s in an inert N_2 atmosphere. The channel length and width of the hybrid phototransistor were 10 μm and 9.3 μm respectively.

The transmission electron microscope (TEM) image was obtained using FEI Technai F20 (WinTech Nano-Technology Services). The location and quality of graphene was identified by optical contrast using an optical microscope and Raman spectroscopy. The Raman spectra and micro PL were obtained using confocal Raman imaging system (Witec alpha300) with a 532 nm wavelength laser. The sample for the PL quenching study was prepared on a substrate with $\sim 1.5 \times 1.5 \text{ cm}^2$ area to ensure a uniformly spin coated NC film, and comprised of both the hybrid perovskite-graphene structure, and the pristine NCs. For a fare scrutiny, the micro-PL intensity of pristine NCs within the near vicinity of the hybrid structure was compared. The TRPL of the samples was measured by a streak camera system with an optimal temporal resolution of $\sim 50 \text{ ps}$. A femtosecond laser with an optical wavelength of 400 nm, a pulse width of 100 fs, and a repetition rate of 1000 Hz was used as the excitation source. The decay times were derived by curve fitting the TRPL data using a bi-exponential function. The X-ray diffraction (XRD) of the NCs layer was carried out using Bruker D8 Advance. All the device measurements were done under vacuum ($\sim 10^{-5}$ torr) and the IV characteristics were collected using Keithley 4200 semiconductor characterization system. The monochromatic LED light with the spectra centered at 525 nm (SOLIS 525C from Thorlabs) was used for all the photoresponse studies. The light intensity was measured using a photodetector and was calibrated before all the measurements. PL measurements for the stability study were done in air using Horiba's FluoroMax-4 tabletop fluorometer with an increment of 0.5 nm from 550 nm to 750 nm at an excitation wavelength of 500 nm. Similar setting was maintained for all the measurements.

Results and discussions

Figure 1a presents the TEM image of the NCs which displays cube shape and a particle size of $\sim 7.17 \pm 1.75 \text{ nm}$ (Figure S1). The particle size coincidentally is higher than the exciton Bohr radius for pure CsPbI₃ NCs ($\sim 6 \text{ nm}$)^{38,44} and CsPbBr₃ NCs (3.5 nm), placing our NCs in the weak confinement regime³⁸, establishing a photophysics similar to the bulk material⁴⁵. This means that the formation of excitons can easily contribute to the generation of photocurrent in the NCs-graphene interface. The X-ray diffraction (XRD) pattern of NCs (Figure 1b) concurs with the cubic lattice structure observed in the TEM analysis and clearly evidences the high crystalline nature of the NCs. The occurrence of lattice expansion I⁻ substitution¹ can be observed using the position of (200) reflection which is at a lower angle of 29° as compared to

pure CsPbBr₃ NCs (30.68°). Moreover, a significant peak intensity corresponding to the (200) plane suggests that the crystals are grown along that plane after the ion exchange reaction¹. Additionally, the shoulder peaks along the low angle reflection in the (111) and (200) directions are likely to be ascribed to organic moieties bound to the NC surface⁴⁶.

Figure 1c presents the ground state absorption of the NCs exhibiting a small absorption band at low energy at 625 nm wavelength probably due to the inter-band absorption background⁴⁴. The perovskite NCs exhibit the absorption band edge at ~650 nm, leading to a higher band gap energy of 1.93 eV, due to quantum confinement effect. The NCs when excited with 575 nm photons yield a photoluminescence spectrum centred at the wavelength corresponding the absorption edge. This is clearly indicative of the mixed halide composition because the emission corresponding to pure CsPbBr₃ and pure CsPbI₃ nanocrystals are centred around 515 nm and 693 nm respectively^{43,47,48}. In addition, with increasing iodine exchange reaction, the PL peak of CsPbBr₃ nanocrystal has been shown to redshift towards the pure CsPbI₃ nanocrystal^{1,43}. The deposited NCs also exhibited good stability when stored under vacuum up to two weeks (Figure 1d).

Figure 1. (a) TEM image of the anion exchanged $\text{CsPbBr}_x\text{I}_{3-x}$ NCs. Inset shows the magnified image of a single NC (b) Thin film XRD of the NCs (c) Steady state absorption and PL spectra of the $\text{CsPbBr}_x\text{I}_{3-x}$ NCs. Inset shows the Tauc plot derived from the absorption spectra (d) Evolution of thin film PL spectra of the NCs over two weeks.

Figure 2a shows the schematic of the hybrid phototransistor with monolayer graphene as the charge transport layer and perovskite NCs as the photo-absorption layer. The quality of the graphene is examined using the Raman spectra. The Raman spectra of graphene layer exhibit (Figure 2b) Lorentzian shaped 2D and G peaks. The absence of D peak suggests a high-quality defect-free graphene layer. The spectra exhibited 2D band and G band respectively centred at 2670 cm^{-1} and 1582 cm^{-1} in agreement to typical Raman spectrum of graphene Si/SiO₂ wafer. Moreover, the $I_{2D}:I_G$ ratio exceeds 2.5, proving the monolayer nature of the graphene, in agreement with the measured thickness of $\sim 0.503\text{ nm}$ obtained from the AFM thickness profile (Figure S2). The work function difference between the NCs and graphene can provide a high potential difference leading to an efficient injection of holes into the monolayer graphene. As such, a qualitative measure of the charge transfer is demonstrated in Figure 2c which shows a significant reduction in quantum yield ($\sim 37\%$ reduction) for the hybrid graphene-NC system, reflecting efficient charge transfer from NCs to graphene layer. Time-resolved PL (TRPL) dynamics are also conducted to obtain a quantitative analysis of the charge injection and separation (see Figure 2d and Table S1). When excited at 532 nm , the fluorescence lifetime of the hybrid NCs-graphene layer is significantly shorter, with an average lifetime of 0.7 ns , as compared to that of pristine NCs layer ($\sim 6.07\text{ ns}$). The considerably low PL lifetime in the hybrid structure demonstrates a substantial charge extraction and transfer from NCs to graphene and thus facilitating an efficient photoresponse.

Figure 2. (a) Schematic of the hybrid phototransistor with the inset showing the crystal structure of NCs with Cs (blue), Pb (yellow), and Br/I (pink), (b) Raman spectra of the pristine graphene monolayer and comparison of the 2D band G band Raman shifts observed in the hybrid structure, indicating the doping level of graphene. (c) Steady-state PL quenching observed in the hybrid structure (d) TRPL spectra showing excellent charge injection into the graphene layer.

The pristine monolayer graphene is found to be slightly n-doped from the transfer curve of the device (Figure 3a). This is further affirmed by the location of 2D band at 2670 cm^{-1} in the Raman spectrum (Figure 2b) which is red shifted from the position commonly observed for undoped graphene on Si/SiO₂ wafer⁴⁹. Upon contact, electron is depleted from the perovskite NCs, further n-doping the graphene, causing an upward band bending within the NCs and rising the Fermi level of graphene. Thus, this doping effect is observed as a shift in Dirac voltage (V_{Dir}) to a lower gate voltage in the transfer curve. Concurrently, both the G band and 2D band in the hybrid structure is found to have red shifted at low light intensity in the Raman spectrum. A red shift in the G band could mean either of the n or p doping effect. However, the red shift in the 2D band is noticeable only for the electron doping effect of perovskite^{50–52}. Under illumination, photon is absorbed by the NCs to generate electron hole pairs (EHP)⁵³. However, owing to the upward band bending in the NCs, hole injection is favoured over the electron injection. Thus, the V_{Dir} is shifted towards higher gate voltage and a corresponding blue shift

in the 2D band is observed in the Raman spectrum establishing a p-doping effect of graphene due to hole injection^{54,55}. Further, we rule out the strain effects on the position of 2D band as that will contribute to significantly larger shift⁴⁹. Figure 3b also illustrates the band structure of the hybrid device before and after graphene-perovskite contact.

Next, we performed a detailed investigation on the phototransistor performance. The transfer curve of the pristine graphene device as well as the hybrid device under dark show a characteristic bell curve of graphene device⁵⁶, proving the dominant charge transport contributed by the graphene channel. For the photoresponse evaluation, the devices are illuminated with a 525 nm monochromatic light. The hybrid device shows higher photocurrent under illumination and exhibits an evident positive shift in Dirac voltage corresponding to hole injection phenomenon from the NCs into graphene. Since the pristine graphene transistor showed no photosensitive behaviour at this wavelength, the photoresponse of the hybrid device is attributed to the photoabsorption properties of the perovskite NCs. Figure 3c summarizes the transfer response of the hybrid device under various input powers at a constant drain bias (V_{DS}) of 5 mV. It can be observed that the Dirac voltage of the device shifts to higher positive voltages with increasing input light power. With increased light intensity, more EHPs are created leading to a greater injection of holes into the transport layer thereby enhancing the photocurrent⁴. Similarly, the output curves in Figure 3d demonstrate a shift in output curve to higher current values due to enhanced photocurrent with increasing input light power.

We next turn to the photoresponsivity of the device extracted from the transfer curves (Figure 3e) which describes the input-output gain of the photodetector and calculated from the photocurrent described by the equation $R = I_{PH}/P$, where I_{PH} is photocurrent described by $I_{PH} = I_{LIGHT} - I_{DARK}$, and P is the total input light power falling on the active device area. The photodetector exhibits excellent photoresponsivity for both gate bias points at $V_{GS} = -60$ V and $V_{GS} = 60$ V, achieving a maximum of responsivity of 11278.14 A/W and 7118.87 A/W respectively at 80 $\mu\text{W}/\text{cm}^2$ illumination. Another figure of merit of the photodetector called specific detectivity (D^*) was also calculated, assuming shot noise as the major source of noise^{23,57}. Consequently, the detectivity, calculated using the equation^{23,53} $D^* = (RA^{1/2}) / (2|e|I_{DARK})^{1/2}$, where e is the charge of electron, I_{DARK} is the dark current and A is active device area, is found to be 1.17×10^{11} Jones and 6.16×10^{10} Jones at $V_{GS} = -60$ V and $V_{GS} = 60$ V respectively. The effect of increasing V_{DS} on the photoresponsivity is presented in Figure S3b at $V_{GS} = -60$ V. With increasing V_{DS} , the transit time for the injected carrier is

decreased, leading to a higher photocurrent and a higher responsivity. Overall, a nearly linear relation between the photocurrent and the input power is observed in Figure 3f.

Figure 3. (a) Transfer curves showing the Dirac voltage shift of the pristine graphene and hybrid device under dark compared against that of the hybrid device under illumination at 525 nm. (b) Schematic diagram of the band bending and Fermi level shifting in NCs and graphene before and after contact, and hole injection from NCs to graphene under illumination (c) Transfer curve of the hybrid device under various input light power. (d) Output curve of the hybrid device under different illumination power. (e) Dependence of responsivity and detectivity on the input light power. (f) Dependence of photocurrent on input power. All the measurements correspond to an active device area of $124.85 \mu m^2$.

The excellent photo-response of the fabricated device can be attributed to two possible reasons. Firstly, owing to the vertical spatial charge separation across the hybrid interface due to energy level mismatch between the perovskite NCs and graphene monolayer, a built-in potential is formed leading to an upward band bending in the NCs which facilitates charge separation and hole injection across the interface. This increases the electron lifetime, leading to an excellent photoconductive gain. Secondly, the field effect mobility of the pristine graphene device is determined to be $826.62 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, which translates to low transit times for holes across the channel causing the holes in the channel to recirculate numerous times before the electrons in the NCs can recombine³.

Figure 4. (a) Temporal photocurrent response of the hybrid device under different gate biases at $80 \mu\text{W}/\text{cm}^2$ illumination (b) Rise and fall times of the phototransistor. The rise time is defined as the time required by the photocurrent to reach 90% from 10% of the maximum photocurrent (vice versa for the fall time)

An important performance parameter of the photodetector is the temporal response. The rise and fall times of the phototransistor was found to be 273.6 ms and 2.26 s respectively with good repeatability (Figure 4a and 4b). The photoconductive gain described by $G=\tau/t$, where τ is photocarrier life time and t is the transit time, is calculated to be 9.32×10^{10} . The high photoconductive gain further substantiates the potential for light sensing under extremely low power light input. The lifetime of the charge carriers estimated through a single exponential curve fitting yielded a carrier lifetime of 2.7 s which is likely to be ascribed to the electron trapping in the NCs⁴. The high fall time may also be either due to the charges trapped in the deep traps³, and/or due to the ligands limiting the charge injection across the interface²⁴. Additionally, the temporal response asserts the negative and positive photocurrent regions

observed in the transfer curve in Figure 3a due to the shift in Dirac voltage. At $V_{GS} = -50$ V, the device is operated in the positive photocurrent region at $80 \mu\text{W}/\text{cm}^2$ illumination, and negative photocurrent region at $V_{GS} = 50$ V.

Figure 5. (a) Stability of the photocurrent over time under continuous illumination of $800 \mu\text{W}/\text{cm}^2$, (b) Stability of the photocurrent over time under periodic illumination of $800 \mu\text{W}/\text{cm}^2$. Inset shows the PL of the NCs before and after the stability test (c) Slow decay of dark current after light exposure (d) Comparison of dark current before and after the photomeasurements

Ion separation and segregated halide domains in the presence of light have reported by several groups working on thin films of mixed halide perovskites such as $\text{MAPb}(\text{I}/\text{Br})_3$ for photovoltaics^{58–60} and $\text{CsPb}(\text{Br}/\text{I})_3$ nanocrystals for light-emitting diodes^{48,58,61}. It is therefore important to investigate this phenomenon in mixed halide perovskite nanocrystals based photodetectors and its photostability under various light intensities which has also not yet been reported. Figures S4a-c show the PL spectra of the NC thin film on a silicon-silicon dioxide substrate at various high light intensities ($55 - 202.3 \text{ mW}/\text{cm}^2$). The photostability testing is carried out under vacuum condition to negate the effects of humidity. All the samples were

exposed to 525 nm light and the PL was measured in air. As evident, the obvious blueshift and increased full width half maximum (FWHM) in PL are consistent with previous reports⁶¹, explained by ionic migration and phase segregation within the NCs^{39,61–63} and driven by thermodynamic forces under light⁵⁸. In addition, we observed that the NCs undergo severe photobleaching effect with this higher intensity light exposure. Hence, the mixed halide NCs are suitable for photodetecting applications since the desired light intensity regime is typically $1 \mu\text{W}/\text{cm}^2$ to $1 \text{ mW}/\text{cm}^2$ ^{3,4,29,58,63}. Therefore, these phototransistors may be coupled with internet of things to implement smart functionality under indoor lighting conditions.

Finally, the photostability of the device over time is investigated at continuous illumination for 5 hrs and periodic illumination (every 20 minutes) for 37 hrs, at $800 \mu\text{W}/\text{cm}^2$ of light intensity (Figure 5a and 5b). The hybrid device exhibits a good photocurrent retention of up to ~95% under continuous illumination over 5 hrs and up to ~82% under periodic illumination over 37 hrs. The PL spectra of the NCs in the hybrid device after subjecting to illumination was further analysed to verify its stability. There is no significant shift in the PL spectra (inset of Figure 5b), further highlighting the high photostability of the mixed halides NCs under lower light intensities. The gradual reduction in photocurrent may be attributed to the prolonged trapping of charge carriers in the deep traps, leading to an increase in dark current. The prolonged trapping of the induced carriers also lead to persistent photoconductivity (PPC), where the photocurrent decay after the light is switched off is rendered slow (Figure 5c). As presented in Figure S5, when the device is exposed to a pulsing light source, the photocurrent decay is not immediately apparent while pulsing. This is because the dark current decay takes place over extremely high timescales, establishing long charge release times in the hybrid device (Figure S5), thereby increasing the dark current (Figure 5d). The presence of deep traps in NCs and/or in the monolayer graphene can also be evidenced by a low exponential value (0.124) derived from the power law of photocurrent⁵¹ (Figure 3e).

Conclusions

In summary, we report CsPbBr_xI_{3-x} NCs-graphene hybrid photodetector, where the monolayer graphene functions as the channel and the highly luminescent perovskite NCs (emission peak at 650nm) act as the absorber layer. The device exhibits a good responsivity of $1.13 \times 10^4 \text{ A/W}$ and a specific detectivity of $1.17 \times 10^{11} \text{ Jones}$. In particular, an ultrahigh photoconductive gain of 9.32×10^{10} is attained. The TRPL and PL quenching studies revealed excellent charge separation and injection of holes into monolayer graphene. The NCs showed superior stability

for up to 2 weeks and the device demonstrated a photocurrent retention of ~82% after 37 hours. However, the slow photoresponse of the device needs to be addressed. In addition, our studies also clarify the effect of light intensities on the stability of the mixed halide NCs and reveal that the common issue of ion separation and segregated halide domains do not occur under low light intensities. The high gain and good photostability substantiates the potential of mixed halide perovskite NCs for light sensing under extremely low power light input.

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Supporting information

Additional experimental details: TRPL parameter table, NC particle size distribution, AFM profile of graphene, output characteristics of the phototransistor, evolution of thin film PL, pulsing response of the device

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