Broadband phototransistors realised by incorporating a bi-layer perovskite/NIR light absorbing polymer channel

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This document is the authors' final version of the published article.
Link to published article: https://doi.org/10.1039/C8TC06229C

APA Citation

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UV to NIR Light Broadband Phototransistors Enabled by a Bi-layer Organo-metallic Halide Perovskite/NIR Light Absorbing Polymer Channel

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Abstract: The high performing ultra violet (UV) to near infrared (NIR) light broadband phototransistors (PTs) are enabled by incorporating a bi-layer methylammonium lead triiodide (MAPbI$_3$) perovskite/NIR light absorbing diketopyrrolopyrrole-dithienylthieno[3,2-b]thiophene (DPP-DTT) polymer channel. The bi-layer MAPbI$_3$/DPP-DTT channel takes the advantages of (1) complementary absorption and (2) high charge transport efficiency of the two materials. The on- and off-state transfer characteristics of the bi-layer MAPbI$_3$/DPP-DTT channel PTs, in the presence of different sources of UV, visible and NIR light, were analyzed. The bi-layer MAPbI$_3$/DPP-DTT channel PTs possess simultaneously a specific detectivity ($D^*$) of $>10^9$ Jones over the UV to visible light wavelength range and a high $D^*$ of $>10^7$ Jones over the NIR light wavelength range. The broadband PTs can be operated at a low voltage (-1 V) without showing the persistent photoconductivity behavior. The results are very encouraging. It is anticipated that the bi-layer perovskite/NIR light absorbing polymer channel concept is a very promising approach for attaining high performance UV to NIR light broadband PTs.
1. Introduction

High-sensitivity solution-processable ultra violet (UV) to near infrared (NIR) light broadband photodetectors are crucial for applications in imaging formation, and safety and wellness monitoring\textsuperscript{1–3}. In comparison to the conventional inorganic semiconductor technologies, solution-processable UV-NIR light broadband photodetectors offer a promising alternative due to their low cost, flexible design, adaptability to various fabrications, and large area manufacturability. The phototransistors (PTs) enable tunable photoresponsivity, high gain by adjusting the gate voltage, and can be operated either electrically or optically for light detection and imaging applications\textsuperscript{4–6}. The performance of the conventional broadband PTs with a single layer channel is often limited due to its narrow absorption spectrum\textsuperscript{7,8} and the imperfect charge transport properties\textsuperscript{6,8}. PTs with a bi-layer heterojunction or a bulk heterojunction (BHJ) channel have some advantages, such as multispectral response\textsuperscript{8}, enhanced responsivity\textsuperscript{9–11} and improved response speed\textsuperscript{6,12}. Various approaches have taken this route, such as incorporating different materials and BHJ channel to improve charge separation processes in the channel\textsuperscript{6,8–12}. However, all had only limited success due to the technical challenges.

The methylammonium lead triiodide (MAPbI$_3$)-based perovskite semiconductors have been adopted for making high efficiency solar cells, achieving power conversion efficiency of $>$20\%\textsuperscript{13–15}. The MAPbI$_3$ perovskite also is a promising material for application in UV-visible light detection. The perovskite-based broadband PTs that incorporated different functional materials including graphene\textsuperscript{16–18}, zinc oxide nanowire\textsuperscript{19}, indium gallium zinc oxide\textsuperscript{20}, and organic materials\textsuperscript{21,22} were reported. However, these hybrid broadband photodetectors have some limitations, e.g., high operating voltage\textsuperscript{21}, sophisticated fabricating procedures\textsuperscript{16–19} and unsatisfied solution fabrication processes\textsuperscript{21}. 
In this work, we propose a novel approach of incorporating a bi-layer MAPbI$_3$/NIR light absorbing diketopyrrolopyrrole-dithienylthieno[3,2-b]thiophene (DPP-DTT) channel for attaining a solution-processable high performance UV-NIR light broadband PT. DPP-DTT polymer has a high NIR absorption and a carrier mobility suitable for high sensitivity NIR light detection. Recently, we demonstrated a high performing solution-processable NIR to visible light up-conversion device, comprising a DPP-DTT/nonfullerene acceptor BHJ NIR charge generation layer and a perovskite light-emitting emitter, for visualizing the NIR light$^1$. The DPP-DTT polymer also is a good candidate for making high performance organic field effect transistors$^{23-25}$, demonstrating high-sensitivity NIR PTs with both high photoresponsivity and photosensitivity$^4$.

The PTs with a bi-layer MAPbI$_3$/DPP-DTT channel in this work take the advantages of (1) complementary absorption of MAPbI$_3$ and DPP-DTT to realize UV-NIR light broadband photodetection, and (2) high charge transport efficiency. The broadband PTs having a specific detectivity ($D^*$) of $>10^9$ Jones over the UV-visible light wavelength range, and a high $D^*$ of $>10^7$ Jones over the NIR light wavelength range were demonstrated. The photoresponsivity enhancement in the bi-layer MAPbI$_3$/DPP-DTT channel PTs at the on- and off-states was analyzed. The solution-processable PT technology thus developed provides the functional superiority for application to large area UV-NIR light broadband photo-detection.

2. Experimental section

The bottom-gate bottom-contact (BGBC) PTs having a bi-layer MAPbI$_3$/DPP-DTT channel were fabricated using the heavily p-doped Si substrates, with a ~300 nm thick thermally grown SiO$_2$ layer acting as the gate dielectric. The Si substrates were cleaned by ultrasonication sequentially
with deionized water, acetone, and isopropyl alcohol each for 15 min. The substrates were dried with a stream of pure N₂ gas and stored in the oven, the SiO₂ surfaces were modified with trichloro (octadecyl) silane (Sigma Aldrich) for the passivation of surface traps and improvement of surface hydrophobicity. 40 nm thick Au source and drain contacts, defining a channel length of 80 μm and a channel width of 1500 μm, were deposited on the modified SiO₂ by thermal evaporation. Then, the DPP-DTT polymer, dissolved in 1, 2-Dichlorobenzene at a concentration of 6 mg/mL, was overlaid on the surface of the BGBC PT template by spin-coating at a rotation speed of 1000 rpm for 240 s in the glovebox, with the oxygen and moisture levels less than 0.1 ppm. After the annealing process at 180 °C for 10 min, the surface of the DPP-DTT polymer layer was modified by the argon (Ar) plasma treatment to create a hydrophilic surface assisting in the growth of the subsequent perovskite layer. The adduct method was employed for preparing the MAPbI₃ layer on DPP-DTT surface. The perovskite precursor solution with a concentration of 1 mole/L was prepared by mixing 1 millimole lead iodide (Xian Polymer), 1 millimole methanaminium iodide (Dyenamo) and 1 millimole dimethyl sulfoxide (Sigma Aldrich) in 1 mL dimethylformamide (Sigma Aldrich). The precursor solution was stirred at room temperature overnight before use. The control PT with a single layer DPP-DTT channel was also fabricated for comparison studies.

The thicknesses of the DPP-DTT layer and perovskite layer were controlled at about 70 nm and 230 nm, respectively, measured using a Veeco Dektak 150 Profilometer. The absorption spectra of the films were recorded using an HP 8453 spectrophotometer. XRD spectra of the perovskite films were measured using a Bruker D8 Advance system (Cu X-ray source). The morphological images of the samples were measured using a LEO 1530 SEM. Current–voltage characteristics of the PTs were measured using a Keithley 2636B dual-channel source meter.
coupled with a probe station. The broadband photoresponses of the PTs were measured using UV (365 nm), visible (450 nm) and NIR (850 nm) LED (Zolix) light sources. The intensity of different light sources was controlled using the neutral optical filters and calibrated using a power meter (Thorlabs). All the measurements were performed in ambient.

3. Results and discussion

The molecular structures of MAPbI$_3$ perovskite semiconductor and DPP-DTT polymer are shown in Fig. 1a and b respectively. The schematic configuration of a BGC type PT with a bi-layer MAPbI$_3$/DPP-DTT channel is shown in Fig. 1c. The absorption spectra measured for the MAPbI$_3$, NIR light absorbing DPP-DTT polymer and bi-layer MAPbI$_3$/DPP-DTT layers are shown in Fig. 1d. Complementary absorption of the bi-layer MAPbI$_3$/DPP-DTT layer over the UV-NIR light wavelength range is clearly seen. The MAPbI$_3$ layer exhibits a good light absorption in the UV-visible light wavelength region. The DPP-DTT also has a high absorption coefficient of $\sim$2.6x$10^5$ cm$^{-1}$, which is much higher than silicon ($\sim$1x$10^3$ cm$^{-1}$) at this wavelength. The absorption of the DPP-DTT polymer layer peaks at 820 nm, with the absorption extending to the long wavelength of 950 nm. Therefore, the use of a bi-layer MAPbI$_3$/DPP-DTT configuration takes the advantage of the strong light absorption over the UV-visible region by MAPbI$_3$ and high NIR absorption by DPP-DTT polymer, making it promising for UV-NIR light broadband photo-detection.

The DPP-DTT polymer layer has a hydrophobic surface, as revealed in the contact angle measurements. The surface of a pristine DPP-DTT polymer layer has a large contact angle of 105°, as shown in Fig. 2a. The hydrophobic polymer surface is unfavorable for the growth of the MAPbI$_3$ layer. To resolve with this problem, the hydrophobic DPP-DTT polymer surface was
modified by the Ar plasma treatment to form a hydrophilic surface. The contact angle measured for the Ar-plasma treated DPP-DTT polymer layer surface decreased dramatically as compared to that measured for the pristine DPP-DTT polymer surface, as shown in Fig. 2b-d, indicating the decrease in the hydrophobicity. The contact angle reduced from 105° for un-treated DPP-DTT surface to 63.1°, 61.5° and 52.7° respectively after 3 s, 5 s and 10 s Ar plasma treatments. It was found that, a 3 s Ar plasma-treated DPP-DTT layer was effective to create a desired surface property for the growth of the MAPbI₃ layer.

The morphological and structural properties of the MAPbI₃ thin films grown on the SiO₂/Si and DPP-DTT surfaces were analyzed using the scanning electron microscopy (SEM) and X-ray diffraction (XRD) measurements. The cross-sectional view and top view SEM images measured for the MAPbI₃ film grown on the SiO₂/Si substrate are shown in Fig. 3a and b, and that measured for a bi-layer MAPbI₃/DPP-DTT film formed on the SiO₂/Si are shown in Fig. 3c and d. The SEM images, shown in Fig. 3c, reveal that a dense and uniform MAPbI₃ layer was formed on the surface of the Ar plasma-treated DPP-DTT polymer layer. There is no observable change in the top view SEM images measured for the MAPbI₃ layers grown on the SiO₂ and DPP-DTT polymer surfaces, suggesting that the MAPbI₃ layers grown on the SiO₂ and DPP-DTT polymer surfaces had similar polycrystalline characteristics. The XRD spectra, as shown in Fig. 3e and f, agree well with the SEM observation in showing that the MAPbI₃ layers, grown on the SiO₂ and DPP-DTT polymer surfaces, possess the same crystallinity and the film quality. It is clear that the XRD patterns measured for the MAPbI₃ layers having three characteristic diffraction peaks at 14.2°, 28.5° and 31.9°, associated with (110), (220) and (310) planes, agree well with the reported results²⁶. These results suggest that the Ar plasma-treated DPP-DTT polymer surface is suitable for the growth of the MAPbI₃ layer.
The transfer curves of the PTs as a function of light intensity were measured. The gate voltage ($V_{GS}$) was scanned from 10 V to -10 V and source-drain voltage ($V_{DS}$) was fixed at -1 V in the measurements. The photoresponses of the broadband PTs were characterized using different UV (365 nm), visible (450 nm) and NIR (850 nm) light sources. The transfer curves measured for a control PT with a single layer DPP-DTT channel using different intensities of the UV, visible and NIR light sources are plotted in Fig. 4a-c. The corresponding transfer curves measured for the broadband PTs with a bi-layer MAPbI$_3$/DPP-DTT channel using different intensities of the UV, visible and NIR light sources are plotted in Fig. 4d-f. The results reveal clearly that the PTs with a bi-layer MAPbI$_3$/DPP-DTT channel exhibit much profound change in the photocurrent, particularly over the UV-visible light wavelength range, as compared to that of the control PT with a single layer channel, implying an enhanced photoresponse.

The photoresponsivity ($R$) of the PTs can be calculated using the equation $R = (I_{light} - I_{dark})/P_{incident}$, where $I_{light}$ and $I_{dark}$ are the channel currents measured for the PTs in the presence of light and the absence of light, respectively. $P_{incident}$ is the intensity of the incidence of light. The values of $R$ were extracted from the transfer characteristics. Photoresponsivity characteristics as a function of $V_{GS}$, measured for the PTs with a single DPP-DTT channel and a bi-layer MAPbI$_3$/DPP-DTT channel using different intensities of the UV, visible and NIR light sources are presented in Fig. 5. It is clear that $R$ of the PTs is dependent on $V_{GS}$, indicating that higher responsivity and gain can be readily obtained by applying an appropriate $V_{GS}$. Photoresponsivities measured for a control PT with a single layer DPP-DTT channel using different intensities of the UV, visible and NIR light sources are shown in Fig. 5a-c. The corresponding photoresponsivities measured for the PTs with a bi-layer MAPbI$_3$/DPP-DTT channel using different intensities of the UV, visible and NIR light sources are plotted in Fig. 5d-


The photoresponsivity of the PTs with a bi-layer MAPbI₃/DPP-DTT channel obviously outperformed that of a control PT with a single layer DPP-DTT channel, particularly in the UV-visible light wavelength region. The results reveal that the PTs with a bi-layer MAPbI₃/DPP-DTT channel possess a clear enhanced UV-NIR light broadband photoresponses, taking the advantages of the complementary absorption of the MAPbI₃ and DPP-DTT, enabled by the bi-layer channel configuration, and thereby leading to a high performance broadband photoresponsivity.

Photoresponsivities measured for a control PT with a single layer DPP-DTT channel and a bi-layer MAPbI₃/DPP-DTT channel, operated under the on-state ($V_{GS} = -8$ V) and off-state ($V_{GS} = 8$ V), as a function of the intensity of UV, visible and NIR light sources are plotted in Fig. 6. The $V_{DS}$ was set at -1 V in the measurements. Photoresponsivities of different PTs operated at the on-state ($V_{GS} = -8$ V) in the presence of UV (2 $\mu$W/cm²), visible 450 nm (2 $\mu$W/cm²) and NIR 850 nm (215 $\mu$W/cm²) light sources are plotted in Fig. 6a, while the corresponding photoresponsivities of the PTs operated at the off-state ($V_{GS} = 8$ V) are presented in Fig. 6b. The results in Fig. 6 reveal clearly that the photoresponsivity enhancement in PTs with a bi-layer channel is realized, operated at the on- and off-states in the presence of UV-visible light. The photoresponsivity values of the PTs with a bi-layer MAPbI₃/DPP-DTT channel, operated at the on-state in the presence of lower light intensity of the UV, visible and NIR light sources, were 7.6 A/W, 9.5 A/W and 0.08 A/W, which are 5, 5.9, and 1.3 times higher than that of a control PT with a single layer DPP-DTT channel measured under the same conditions in the presence of UV light (1.5 A/W), visible light (1.6 A/W), and NIR light (0.06 A/W). At the off-state, the PTs with a bi-layer MAPbI₃/DPP-DTT channel had photoresponsivities of 0.38 A/W, 0.43 A/W and 0.0023 A/W in the presence of UV, visible and NIR light sources, which are 214, 480 and 230.
times higher than that measured for the PTs with a single layer polymer channel under UV
(1.77×10^{-3} A/W), visible (8.9×10^{-4} A/W) and NIR (1×10^{-5} A/W) light sources, respectively. The
significant broadband enhancement in the photoresponsivity is enabled by incorporating a bi-
layer MAPbI₃/DPP-DTT channel in the PTs, taking the advantages of (1) complementary
absorption of MAPbI₃ and DPP-DTT, and (2) high charge transport efficiency in the perovskite
in the MAPbI₃/polymer channel.

To unravel the origin of the advantages of the bi-layer MAPbI₃/DPP-DTT channel on the
photoresponsivity enhancement in the broadband PTs, the transistor properties of both types of
the PTs in the absence of light were examined. The dark transfer curves as a function of gate
voltage and source-drain voltage, measured for the PTs with a bi-layer MAPbI₃/DPP-DTT
channel under different V_DS in the dark, are plotted in Fig. 7a, exhibiting typical p-type
conduction behavior. It shows that, by varying the V_DS from -1 V to -10 V, the change in channel
current (I_DS) in the PTs operated at the off-state is more prominent than the change in I_DS
measured for the PTs at the on-state. At the off-state, the I_DS measured for the PTs with a bi-layer
MAPbI₃/DPP-DTT channel was higher than that of a control PT with a single layer DPP-DTT
channel, operated at V_DS= -1 V, as shown in Fig. 7b. While at the on-state, both types of PTs
exhibit similar transistor behavior with comparable I_DS, as shown in Fig. 7b. At the on-state, the
charge carrier transport preferably though the DPP-DTT layer due to the accumulation of the
holes at the DPP-DTT/SiO₂ interface, induced by a negative V_{GS}, forming an ultra-high
conductance of the polymer channel. At the off-state, no holes are accumulated near the vicinity
of the DPP-DTT/SiO₂ interface in the bi-layer MAPbI₃/DPP-DTT channel. In such a situation, a
relatively higher I_DS, observed in the PTs with a bi-layer MAPbI₃/DPP-DTT channel at the off-
state, comes from the transport of the charge carriers in the perovskite layer. To illustrate the
point, $I_{DS}$, measured for the PTs with a bi-layer MAPbI$_3$/DPP-DTT channel and a single layer DPP-DTT channel at the off-state ($V_{GS}=8$ V), as a function of $V_{DS}$ were measured. $I_{DS}$, measured for a PT with a single layer MAPbI$_3$ channel at $V_{GS}=0$ V, as a function of $V_{DS}$ also was presented for comparison. The results are presented in **Fig. 7c**. With the increase in $V_{DS}$, it shows clearly that $I_{DS}$, measured for the PTs with a bi-layer MAPbI$_3$/DPP-DTT channel, is much higher than that measured for a control PT with a single layer DPP-DTT channel. While $I_{DS}$ measured for the PTs with a single layer MAPbI$_3$ channel is similar to the one observed for the PTs with a bi-layer MAPbI$_3$/DPP-DTT channel. Our results reveal clearly that the higher $I_{DS}$ seen in the PTs with a bi-layer MAPbI$_3$/DPP-DTT channel, operated at the off-state, is enabled primarily by the hole transport in the MAPbI$_3$ layer, which has a higher hole mobility as compared to that in the DPP-DTT layer. Apparently, the hole transport in the DPP-DTT layer was hindered in the PTs operated at a positive $V_{GS}$. The schematic diagrams illustrating the device structures and the current flows in the PTs with a single layer DPP-DTT channel, a bi-layer MAPbI$_3$/DPP-DTT channel, and a single layer MAPbI$_3$ channel are presented in **Fig. 7d-f**.

PTs with a bi-layer MAPbI$_3$/DPP-DTT channel are able to provide two charge transport paths by varying the $V_{GS}$: (1) the MAPbI$_3$ charge transport path for the PTs operating at the off-state, and (2) the DPP-DTT charge transport path for the PTs operating at the on-state. The unique feature of dual charge transport path, enabled by the bi-layer MAPbI$_3$/DPP-DTT channel, offers an attractive photo detection option for the broadband PTs that can be operated at both the off-state and on-state for enhanced photoresponsivity. The photogenerated excitons in the MAPbI$_3$ perovskite layer can be dissociated efficiently due the low exciton binding energy$^{27}$, generating a high density of the charge carriers in the presence of light, which can be efficiently transported in the MAPbI$_3$ layer at the off-state and in the DPP-DTT layer at the on-state. The
schematic diagrams illustrating the current flows in the PTs with a bi-layer MAPbI\(_3\)/DPP-DTT channel at the off-state and on-state in the presence of UV-visible light are shown in Fig. 8a and b. At the off-state, the PTs with a bi-layer MAPbI\(_3\)/DPP-DTT channel possess a higher photoresponsivity due to the strong light absorption in the UV-visible light range and excellent electrical properties of the MAPbI\(_3\) layer\(^{28,29}\) as compared to that of a control PT with a single layer DPP-DTT channel. At the on-state, the high density of the photogenerated charge carriers in the MAPbI\(_3\) layer in the bi-layer MAPbI\(_3\)/DPP-DTT channel can be diffused to the underlying DPP-DTT layer, as schematically shown in Fig. 8b. This helps to increase the density of the charge carriers in the DPP-DTT layer, resulting in a higher channel current as compared to that of a control PT with a single layer DPP-DTT channel. Therefore, an enhancement in the photoresponsivity of the PTs with a bi-layer MAPbI\(_3\)/DPP-DTT channel over the UV-visible light region was observed at both the off- and on-states.

The transfer of the charge carriers between the MAPbI\(_3\) layer and DPP-DTT layer was further analyzed using the steady state and time-resolved photoluminescence (PL) measurements. PL characteristics are closely associated with the behavior of the charge transfer process near the vicinity of the interface between two adjacent photosensitive materials,\(^{30}\) providing a qualitative information of charge transfer and recombination processes. The steady state and time-resolved PL spectra measured for the single layer MAPbI\(_3\) film and bi-layer MAPbI\(_3\)/DPP-DTT film formed on the SiO\(_2\)/Si substrates were measured. The results are plotted in Fig. 8c and d. There is an obvious decrease in the intensity of the steady state PL spectra measured for the bi-layer MAPbI\(_3\)/DPP-DTT as compared to that measured for the single layer MAPbI\(_3\) film over the wavelength range from 650 nm to 850 nm, revealing the processes of the charge transfer from MAPbI\(_3\) layer to DPP-DTT layer taking placing at the MAPbI\(_3\)/DPP-DTT interface. The time-
resolved PL characteristics indicate that photogenerated charge carriers with a shorter lifetime were observed in the MAPbI$_3$ when it was formed on DPP-DTT surface, caused by the charge transfer occurred at the MAPbI$_3$/DPP-DTT interface. Based on these PL results, it is anticipated that the charge transfer occurred at the MAPbI$_3$/DPP-DTT interface in the broadband PTs, is responsible for the photoresponsivity enhancement.

The photosensitivity ($P$) of the different PTs, defined as the ratio of the photocurrent to the dark current, was analyzed. Photosensitivity characteristics as a function of $V_{GS}$, measured for a control PT with a single layer DPP-DTT channel using different intensities of the UV, visible and NIR light sources are presented in Fig. 9a-c. The photosensitivity as a function of $V_{GS}$, measured for the PTs with a bi-layer MAPbI$_3$/DPP-DTT channel using different intensities of the UV, visible and NIR light sources are plotted in Fig. 9d-f. The PTs with a bi-layer MAPbI$_3$/DPP-DTT channel possess a high photosensitivity, which are clearly demonstrated as compared to that of a control PT with a single layer DPP-DTT channel over the UV-visible light region. The observed enhancement in the photosensitivity of the PTs with a bi-layer channel also is associated with the high absorption in the MAPbI$_3$ layer over the UV-visible light region.

The advantage of the broadband PTs with a bi-layer MAPbI$_3$/DPP-DTT channel can be further elucidated by analyzing $D^*$ of the PTs. Shot noise of a photo detector can be described by $(\sqrt{2qI_{dark}\Delta f})$, where $q$ is the electron charge, $I_{dark}$ is the dark current, and $\Delta f$ is the bandwidth. Shot noise is frequency independent, such that the specific detectivity can be estimated using the equation: $D^* = (R\sqrt{A})/\sqrt{2qI_{dark}}$, where $R$ is the photoresponsivity, and $A$ is the active area of the detector. The wavelength-dependent specific detectivity of the PTs as a function of light intensity was analyzed. $D^*-V_{GS}$ characteristics, measured for a control PT with a single layer DPP-DTT channel using different intensities of UV, visible and NIR light sources.
are shown in Fig. 10a-c. The corresponding $D^* - V_{GS}$ characteristics, measured for the PTs with a bi-layer MAPbI$_3$/DPP-DTT channel using different intensities of UV, visible and NIR light sources are plotted in Fig. 10d-f. Benefiting from the much enhanced photoresponsivity in the UV-visible region, $D^*$ of the PTs with a bi-layer MAPbI$_3$/DPP-DTT channel was much higher than that of a control PT with a single layer DPP-DTT PT channel.

The transient photoresponse speed of the PTs was measured using a 0.5 Hz modulated visible light (450 nm) source with an intensity of 2 $\mu$W/cm$^2$. $V_{GS}$ and $V_{DS}$ used in the transient measurements were fixed at -1 V. The results are shown in Fig. 11a. It is clear that the photoresponse speed measured for the PTs with a bi-layer MAPbI$_3$/DPP-DTT channel was much faster than that of a control PT with a single layer DPP-DTT channel. The rise and fall time of the PTs are estimated by measuring the time taken for photo current rising from 10% to 90% of its maximum value and dropping from the 90% to 10% of its maximum value, respectively. The results are plotted in Fig. 11b. The broadband PTs with a bi-layer MAPbI$_3$/DPP-DTT channel have a much fast response time, e.g., rising time of 0.17 s and falling time 0.25 s, than that of a control PT with a single layer DPP-DTT channel, (e.g., >2.0 s) for both rising and falling time. The results consist with the observation reported in our previously work$^4$. The slow photoresponse obtained for the control PT with a single layer DPP-DTT channel is associated with the presence of defeats, relatively short exciton diffusion length$^{31}$, and imperfection of the charge separation and transport in the single layer channel$^{32}$.

The performance of all polymer-based PTs is often limited due to the low carrier mobility and humble absorption over a broader spectral range. There are not many choices in the photoactive polymer materials that possess simultaneously high NIR absorption and high carrier mobility. The relatively low photoresponsivity and photosensitivity in solution-processable
broadband polymer PTs are still an open challenge. The accomplishment of broadband PTs with a bi-layer perovskite/NIR light absorbing polymer channel as demonstrated in this work will have a profound impact on the development of high performance broadband photodetectors.

4. Conclusions
In conclusion, the high performing UV to NIR light broadband PTs with a bi-layer MAPbI$_3$/DPP-DTT channel was demonstrated. The use of a bi-layer MAPbI$_3$/DPP-DTT channel largely enhances the photoresponsivity, photosensitivity and specific detectivity of the broadband PTs. The bi-layer MAPbI$_3$/DPP-DTT channel PTs also possess enhanced transient photoresponses as compared to that of a control PT with a single layer DPP-DTT channel, benefiting from the combined effects of complementary absorption and high charge transport in the bilayer channel. It is anticipated that the bi-layer channel PT developed in this work is very attractive for application in low energy consumption and portable electronics.

Conflicts of interest
There are no conflicts to declare.

Acknowledgments
This work was financially supported by the Research Grants Council of Hong Kong Special Administrative Region, China, General Research Fund (GRF/12302817) and Hong Kong Baptist University Inter-institutional Collaborative Research Scheme (RC-ICRS/15-16/04).
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Fig. 1 Molecular structures of (a) MAPbI₃ perovskite and (b) DPP-DTT polymer semiconductors. (c) The schematic cross-sectional view of a PT with a bi-layer MAPbI₃/DPP-DTT channel. (d) The absorption spectra measured for a MAPbI₃ layer, a DPP-DTT layer, and a bi-layer MAPbI₃/DPP-DTT film.
Fig. 2 Contact angles measured for surfaces of the (a) pristine DPP-DTT polymer layer, and the DPP-DTT polymer layers modified by the Ar plasma treatment for (b) 3 s, (c) 5 s and (d) 10 s, respectively.
Fig. 3 (a) Cross-sectional view and (b) top view SEM images measured for a MAPbI$_3$ layer grown on SiO$_2$/Si substrates. (c) Cross-sectional view and (d) top view SEM images measured for a MAPbI$_3$ layer grown on the surface of plasma-treated DPP-DTT layer. XRD patterns measured for the MAPbI$_3$ films grown on (e) SiO$_2$ and (f) Ar plasma-treated DPP-DTT surfaces.
Fig. 4 Transfer curves measured for a control PT with a single layer DPP-DTT PT channel using different intensities of (a) UV, (b) visible and (c) NIR light sources. The corresponding transfer curves measured for a broadband PT with a bi-layer MAPbI$_3$/DPP-DTT channel using different intensities of (d) UV, (e) visible and (f) NIR light sources. The transfer curves were measured at a fixed $V_{DS}$ of -1 V.
**Fig. 5** Photoresponsivity characteristics measured for a control PT with a single layer DPP-DTT channel using different intensities of (a) UV, (b) visible and (c) NIR light sources. The corresponding photoresponsivity characteristics measured for a broadband PT with a bi-layer MAPbI3/DPP-DTT channel using different intensities of (d) UV, (e) visible and (f) NIR light sources. $V_{DS}$ was set at -1 V in the measurements.
Fig. 6 Photoresponsivity characteristics, measured for a control PT with a single layer DPP-DTT channel and a broadband PT with a bi-layer MAPbI$_3$/DPP-DTT channel operated under (a) on-state (at $V_{GS}=-8$ V) and (b) off-state (at $V_{GS}=8$ V), as a function of the intensity of UV, visible and NIR light sources. $V_{DS}$ was set at -1 V in the measurements.
Fig. 7 (a) Transfer characteristics measured for a broadband PT with a bi-layer MAPbI$_3$/DPP-DTT channel under different V$_{DS}$ in the absence of light. (b) Comparison of the transfer curves measured for a broadband PT with a bi-layer MAPbI$_3$/DPP-DTT channel and a control PT with a single layer DPP-DTT channel operated at V$_{DS}$= -1 V. (c) The dark channel currents measured for different PTs at the off-state (V$_{GS}$=8 V), as a function of V$_{DS}$. The dark channel current, measured for a PT with a single layer MAPbI$_3$ channel at V$_{GS}$=0 V, as a function of V$_{DS}$, is also presented for comparison. Schematic diagrams illustrating the device structures and the current flows in the PTs with (d) a single layer DPP-DTT channel and (e) a bi-layer MAPbI$_3$/DPP-DTT channel at the off-state, and (f) a single layer MAPbI$_3$ channel at V$_{GS}$=0V.
Fig. 8 Schematic diagrams illustrating the current flows in a broadband PT with a bi-layer MAPbI₃/DPP-DTT channel at the (a) off-state and (b) on-state in the presence of UV-visible light. (c) Steady state and (d) time-resolved PL characteristics measured for a MAPbI₃ film and a bi-layer MAPbI₃/DPP-DTT film grown on the SiO₂/Si substrates.
Fig. 9 Photosensitivity characteristics as a function of $V_{GS}$, measured for a control PT with a single layer DPP-DTT channel using different intensities of (a) UV, (b) visible and (c) NIR light sources. The corresponding photosensitivity characteristics as a function of $V_{GS}$, measured for a broadband PT with a bi-layer MAPbI$_3$/DPP-DTT channel using different intensities of (d) UV, (e) visible and (f) NIR light sources. $V_{DS}$ was set at -1 V in the measurements.
**Fig. 10** $D^*$–$V_{GS}$ characteristics, measured for a control PT with a single layer DPP-DTT channel using different intensities of (a) UV, (b) visible and (c) NIR light sources. The corresponding $D^*$–$V_{GS}$ characteristics, measured for a PT with a bi-layer MAPbI$_3$/DPP-DTT channel using different intensities of (d) UV, (e) visible and (f) NIR light sources. $V_{DS}$ was set at -1 V in the measurements.
Fig. 11 (a) Transient photoresponses measured for a control PT with a single layer DPP-DTT channel and a broadband PT with a bi-layer MAPbI$_3$/DPP-DTT channel using a 0.5 Hz modulated visible light (450 nm) source with an intensity of 2 $\mu$W/cm$^2$. $V_{GS}$ and $V_{DS}$ were fixed at -1 V. (b) The broadband bilayer channel PTs have a photoresponse time, e.g., a rising time of 0.17 s and a falling time of 0.25 s as compared to a control PT with a DPP-DTT channel (>2.0 s).
Broadband phototransistors (PTs) with a bi-layer MAPbI₃/polymer channel take the advantages of complementary absorption and high charge transport efficiency of the two materials. The broadband PTs possess simultaneously a specific detectivity ($D^*$) of >10⁹ Jones over the wavelength range from UV to visible light and a high $D^*$ of >10⁷ Jones over the NIR light wavelength range.