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Organic and quantum dot hybrid photodetectors: towards full-band and fast detection

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Photodetectors hold great application potential in many fields such as image sensing, night vision, infrared communication and health monitoring. To date, commercial photodetectors mainly rely on inorganic semiconductors, *e.g.*, monocrystalline silicon, germanium, and indium selenide/gallium with complex and costly fabrication, which are hardly compatible with wearable electronics. In contrast, organic conjugated materials provide great superiority in flexibility and stretchability. In this Highlight, the unique properties of organic and quantum dot photodetectors were firstly discussed to reveal the great complementarity of the two technologies. Subsequently, the recent advance of organic/quantum dot hybrid photodetectors was outlined to highlight their great potential in developing broadband and high-performance photodetectors. Moreover, the multiple functions (*e.g.*, dual-band detection and upconversion detection) of hybrid photodetectors were highlighted for their promising application in image sensing and infrared detection. Lastly, we present a forword-looking discussion on the challenges and our insights for the further advancement of hybrid photodetectors. This work may spark enormous research attention in organic/quantum dot electronics and advance the commercial applications.

Introduction

Photodetectors, converting photons to electrical signals, have drawn worldwide research interest due to their versatile applications, including image sensing, spectrometry, optical communication and pulse oximetry.[1–7] Commercially available photodetectors are mainly fabricated with vacuum-processed inorganic semiconductor materials *e.g.*, silicon, indium gallium arsenide and germanium, which generally have complex and costly fabrication.[2,8–11] Moreover, high brittleness also compromises their detection performance and places some restrictions

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on their applications in wearable electronics.[12–16] As promising alternatives to high-cost and rigid photodetectors, solution-based photodetectors can address these issues. Organic photodetectors (OPDs)[3,17,18] and quantum dot photodetectors (QPDs)[19–23] are the two representative technologies, which are compatible with large-scale processing techniques, including blade coating, slot-die coating, spray coating and inkjet printing.

During the last decade, OPDs have witnessed great advances in terms of detection performance and applications. Due to the ease of processing, tailorable response and high flexibility, OPDs have exhibited great application potential in real-time health monitoring, electronic eyes, *etc.*[24–26] Kippelen's group recently demonstrated that OPDs based on polymeric bulk heterojunctions can achieve the comparable detection performance with silicon photodetectors with a detectivity of $\sim 8 \times 10^{12}$ Jones.[27] Nevertheless, the developed OPDs with P3HT:ICBA can only exhibit the confined detection range of $\sim$600 nm, which is significantly narrower than that of silicon photodetectors. With the rise of non-fullerene acceptors, especially for the star Y6 series, high-performance OPD can achieve a high detectivity of over 10^{14} Jones with a broadened detection range of $\sim$950 nm, revealing the great application potential of OPDs for near-infrared (NIR) detection.[28–30] Owing to the intrinsic properties of organic semiconductors, *e.g.*, low carrier mobility and high exciton binding energy, OPDs generally exhibit the low dark current (J_{dark}) of even up to $\sim 10^{-11}$ A cm^{-2}, yet accompanied by lower responsivity and response speed, compared with their inorganic counterparts. Moreover, OPDs generally exhibit the low infrared detectivity of below 10^{11} Jones at the infrared band over 1200 nm, greatly blocking some special applications.

On the other hand, quantum dots (QDs) have emerged as one of the most promising materials for photodetectors, due to their facile absorption tunability, broad response range and high detection performance.[48,58–60] The absorption of QDs can be significantly tuned from X-rays to short-wave infrared (SWIR) lights or even to mid-wave infrared lights (MIR) (Fig. 1a), which can markedly broaden the application of photodetectors, including infrared communication, infrared imaging, remote sensing and night vision.[61–63] For instance, mercury telluride QDs have been reported to exhibit the tunable infrared response from short-wave infrared to mid-wave infrared lights (2500–5000 nm), which can be used for infrared images to identify the temperature difference.[54,64–67] Recently, Guyot-Sionnest's group developed a two-terminal dual-band photodetector based on mercury telluride (HgTe) QDs for bias-switchable spectral response in two distinct bands.[57] The developed photodetector presented the rapid switch between short-wave infrared and mid-wave infrared with a detectivity of above 10^{10} Jones, which can be employed for dual-band imaging and remote temperature recognition. For near infrared and short-wave infrared bands, lead chalcogenide QDs have been widely used to achieve the superior detectivity of over 10^{12} Jones even at $>$1400 nm with high response speed.[50,52,68] Nevertheless, the slightly high J_{dark} of over 10^{-9} A cm^{-2} has placed some restrictions on the further improvement of detection performance for QPDs.

From the above discussion, it can be clearly seen that OPDs and QPDs have demonstrated the great complementarity in detection range and performance (Fig. 1b and Tables 1 and 2). On the one hand, QD materials can be employed to broaden the detection range of OPDs, even to short-wave infrared band.[69] Moreover, the infrared detectivity and response speed of OPDs can be further enhanced with the introduction of QD materials, which will certainly advance the application of OPDs for infrared detection. On the other hand, organic semiconductors can be used to reduce the J_{dark} of QPDs, which can endow a promising chance to further enhance the infrared detectivity to over 10^{13} Jones. Additionally, OPD devices generally employ the donor:acceptor bulk heterojunction for high detection performance, while QPD devices mostly employ the bilayer heterojunction (Fig. 1c). The orthogonally processing solvents for organic and QD materials enable the hybrid strategy of these two materials for high detection performance. With these benefits, organic and QD hybrid photodetectors have witnessed great progress and achieved high detection performance at the broad wavelength ranges. Accordingly, we outlined the recent progress of organic/QD hybrid photodetectors, with the aim of further broadening the response range and enhancing the detection performance. The great complementarity of OPDs and QPDs was highlighted for their great potential to be leveraged for the multi-win. We hold that organic/QD hybrid photodetectors are expected to become the perfect detection devices with the broad detection range, low noise current, high detectivity and fast response speed.

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Organic photodetectors: superior flexibility and great detection performance

Owing to the great superiority of organic semiconductors in wearable electronics, OPDs have witnessed dramatic advances in the last decade. There exist some comprehensive and critical

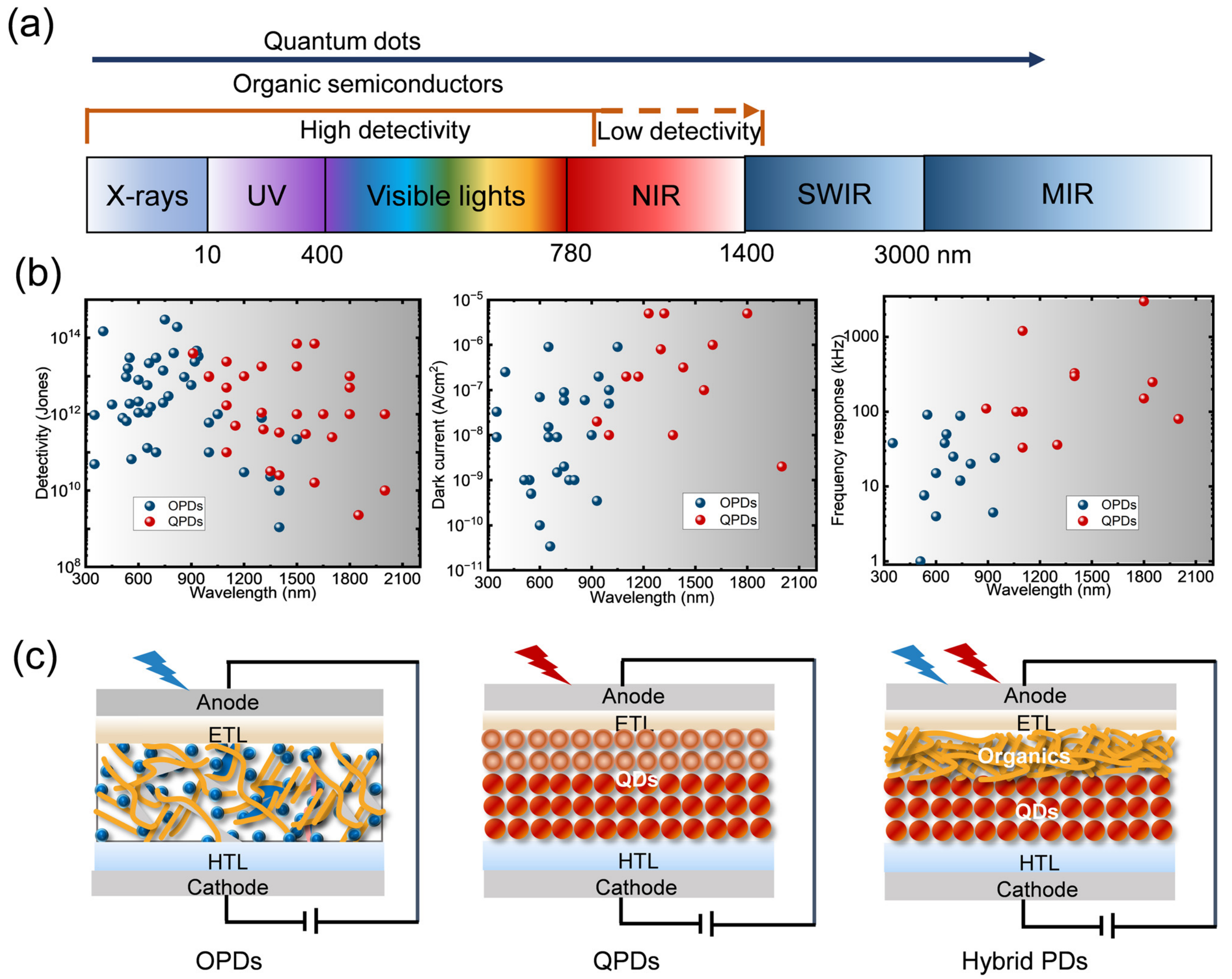

Fig. 1 (a) The response range of organic semiconductors and quantum dots from X-rays to mid-wave infrared lights. Generally, OPD can only high detectivity within 900 nm, and the detectivity will gradually reduce beyond the range. (b) The detection performance comparison between OPDs and QPDs, including detectivity, dark current and response speed. The detection parameters of 30 typical OPDs and QPDs are summarized in Tables 1 and 2. (c) Schematic of the device structures of OPDs, QPDs and hybrid PDs.

Table 1 Summary of the OPD performance in different bands

Wavelength (nm)	D^* (10^{12} Jones)	J_{dark} (10^{-6} A cm^{-2})	LDR (dB)	Response time (μs)	Cut-off frequency (kHz)	Ref.
350	0.95 (−1)	0.009 (−1)	—	—	38	31
550	30 (−1)	5 × 10^{-4} (−1)	148	—	91	32
560	0.066 (−2)	90 (−2)	132	—	864	33
600	2(−1.5)	0.0001 (−1.5)	160	—	15	27
650	1.1 (−1)	0.009 (−1)	170	—	38	31
660	21.9 (−5)	3.4 × 10^{-5} (−5)	160	—	50	34
700	0.1 (−3)	0.00148 (−3)	75	—	350	35
740	14.1	0.059	77.9	2.1	118.3	36
800	40 (−0.1)	0.001 (−0.1)	65.14	—	20	37
900	5.84 (−1)	0.01 (−1)	—	5.03	—	38
940	33.1 (−2)	0.2 (−2)	148	—	240	3
1000	0.6 (−1)	0.1 (−1)	135	7.1	2000	39
1360	10 (−2)	8 × 10^{-4} (−2)	—	—	—	40
1400	0.01 (−2)	900 (−2)	180	—	1000	41
1600	0.03 (−0.1)	1.38 × 10^{-5}	—	—	—	42

reviews on the progress of OPDs from material design, device engineering to applications.[16,70,71] This highlight mainly focused on the advantages of OPDs and also revealed the obstacle for the further performance breakthrough.

Table 2 Summary of the QPD performance in different bands

Wavelength (nm)	D^* (10^{12} Jones)	J_{dark} (10^{-6} A cm^{-2})	LDR (dB)	Response time (μs)	Cut-off frequency (kHz)	Ref.
600	1.71(−10)	—	60.9	3.63	—	43
640	24 (0)	0.001 (0)	—	—	1200	44
790	4.92(−3)	1.0(−3)	105	45.4	7.7	45
920	0.515	—	—	43.0	10	46
980	9.0	—	—	0.7	320	47
1125	0.32(−0.5)	—	—	0.33	—	48
1370	0.7 (−1)	1.0 (−1)	—	0.6	—	49
1550	1.6	—	—	0.007	—	50
1550	1.0 (0)	0.1 (0)	—	—	—	51
1550	0.8 (0)	0.14 (0)	—	0.01	—	52
1550	0.02 (−1)	—	—	110	—	53
1700	0.39 (−0.4)	2.3 (−0.4)	112	6.4	50	54
2200	0.1 (−0.5)	300 (−0.5)	—	—	—	55
2300	0.1 (−12)	—	—	1.5	—	56
5000	0.1 (−0.5)	—	—	2.5	—	57

Note: bias was provided for detectivity and dark current density.

Owing to the great flexibility of organic semiconductors, flexible OPDs have broad applications in wearable electronics for health monitoring, infrared communication and image sensing. Fuentes-Hernandez *et al.* fabricated a 1 cm^2 flexible OPD with a high detectivity of 2.0×10^{12} Jones.[27] The flexible OPD has successfully been used in photoplethysmography (PPG) to evaluate the cardiac and pulmonary functions, which has achieved low signal-to-noise, comparable to that of small-area Si-based photodetectors. Recently, Fang's group developed the ultra-flexible and dual-polarity OPDs by replacing the commonly used ITO/Ag electrodes with the organic electrodes (Fig. 2a).[72] The developed flexible OPDs exhibited the high self-powered responsivity over 0.1 A W^{-1} and can retain ∼80% of the initial performance after bending 20 000 circles. Another promising work by Park *et al.* provided the in-depth investigation of OPD mechanical properties.[73] They developed a stretchable OPD *via* introducing the promising elastomer (SEBS) into the P3HT:ICBA bulk heterojunction, which can enable the low

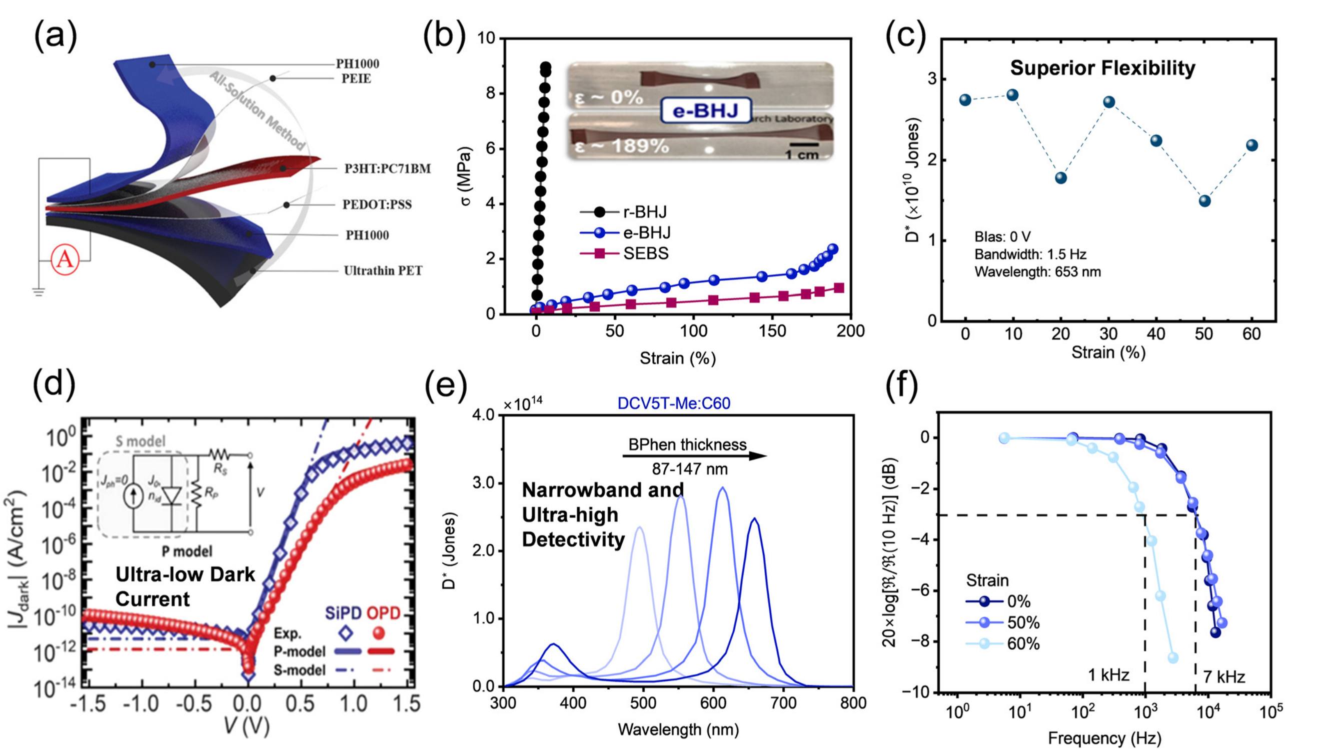

Fig. 2 The properties of OPDs. (a) Schematic of the flexible OPDs. (b) The mechanical behavior of flexible OPDs. (c) The detection performance of the flexible OPDs at different strains. (d) The comparison of dark current between Si photodetectors and OPDs. (e) The detection performance of selective OPDs. (f) The response speed of the flexible OPDs at different strains. (a) Reproduced with permission from ref. 72. Copyright 2022, Wiley. (b, c and f) Reproduced with permission from ref. 73. Copyright 2021, AAAS. (d) Reproduced with permission from ref. 27. Copyright 2020, AAAS. (e) Reproduced with permission from ref. 29. Copyright 2021, Wiley.

Young's modulus approaching the value of human tissues. The developed stretchable OPD can achieve the low J_{dark}, high responsivity and detectivity, even under at least 60% strain, indicating the great superiority of organic semiconductors in developing high-performance stretchable OPDs (Fig. 2b and c). Moving forwards, our group recently investigated in detail the photovoltaic and mechanical properties of polymer:nonfullerene acceptor blends *via* introducing the elastomer SEBS.[74] We found the modulus of the ternary blends can be predicted by a mechanical model, which may also provide a critical guideline for stretchable OPDs.

Apart from the superior flexibility, OPDs also exhibited ultra-low J_{dark} and exceedingly high detectivity, compared to Si-based and other emerging photodetectors. For instance, the pioneering report has demonstrated that diode-based OPDs can achieve the record low J_{dark}, even approaching 10^{-14} A cm^{-2} without bias, which markedly exceeds the values of perovskite and quantum dot photodetectors (Fig. 2d).[27] The ultra-low J_{dark} mainly stems from the intrinsic organic molecular packing and device structure, which can be used to enhance the detection performance of other emerging photodetectors.[29,75] Additionally, OPDs can facilely achieve narrowband detection *via* charge collection narrowing and present ultra-high detectivity. Recently, Xing *et al.* developed a tunable spectral photodetector *via* designing the cavity structure, resulting in narrowband OPDs with the broad band of 400–1100 nm. With this structure, the designed OPDs can present the narrowband wavelength-selective photo-response with a striking detectivity over 10^{14} Jones and a full-width-at-half-maximum of ~40 nm (Fig. 2e). Despite the benefits, OPDs generally exhibit the low response speed with a fall time of tens of milliseconds or a f_{-3dB} of several kHz (Fig. 2f), which are far below that of QD photodetectors with a fall time of tens of nanoseconds. The low response speed may place great restrictions on the applications of OPDs for fast detection, which may be addressed *via* hybrid strategies.

Quantum dot photodetectors: broad detection, fast response and multi-function

QD materials have the facile and extensive tunability of energy level and absorption,[76–79] which outperform most semiconductors in developing high-performance photodetectors covering the broad wavelength range from X-rays to mid-wave infrared lights. The commonly used QD materials in photodetectors mainly include cadmium chalcogenide QDs,[80–83] lead chalcogenide QDs,[84–87] perovskite QDs[88–91] and HgTe QDs,[57,65,67] which nearly meet the detection requirement of various applications. For instance, Kim *et al.* designed the two-dimensionally pixelated full-color photodetector with the monolithic integration of various-sized QDs, including 3 nm CdS QDs for blue-light detection, 5 nm CdSe QDs for green-light detection, 7 nm CdSe QDs for red-light detection and 10 nm PdS QDs for infrared detection (Fig. 3a).[92] The developed full-color photodetectors (phototransistor-based) can enable efficient carrier transport, resulting in the high detectivity

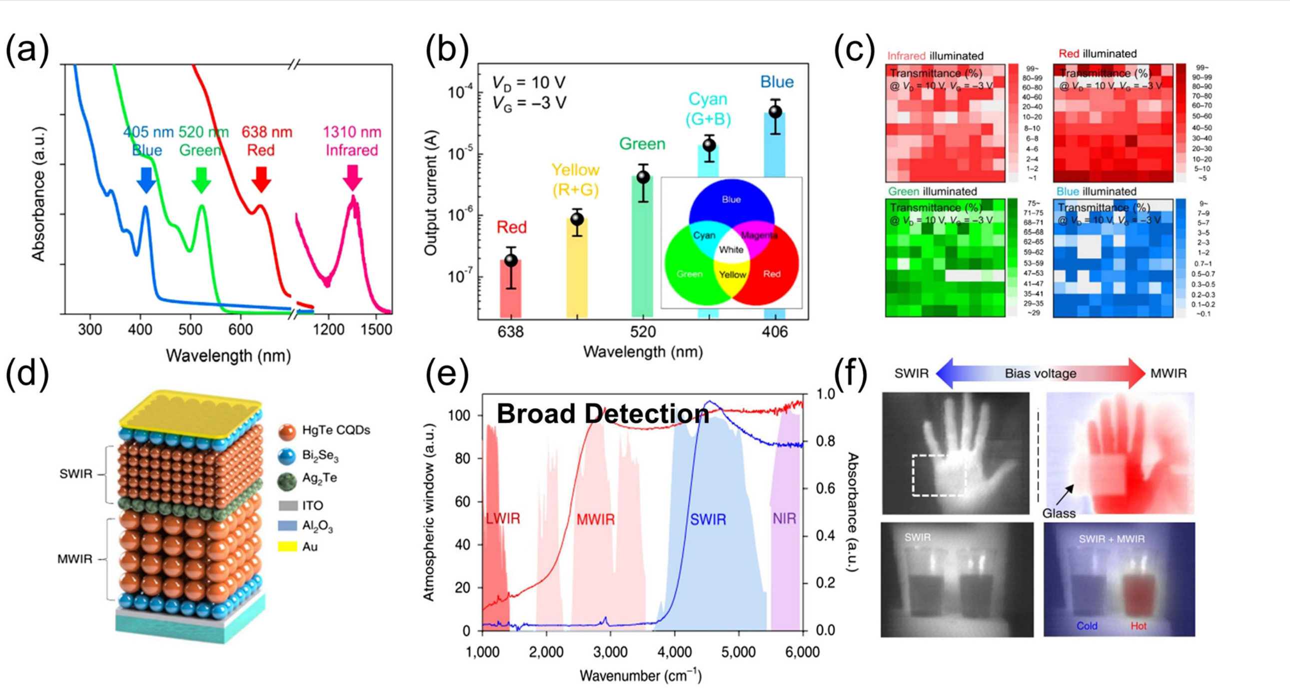

Fig. 3 The properties of QPDs. (a) The absorption of QDs for full-color photodetectors. (b) Output current of the full-color photodetectors. (c) Full-color mapping images of human fingertip. (d) Schematic of the device structure for dual-band infrared imaging. (e) The broad absorption of HgTe QDs. (f) The application of HgTe QDs in dual-band imaging. (a–c) Reproduced with permission from ref. 92. Copyright 2019, Springer Nature. (d–f) Reproduced with permission from ref. 57. Copyright 2019, Springer Nature.

and a responsivity of 8.3×10^3 A W^{-1} in the broad range of wavelengths from 365 to 1310 nm (Fig. 3b). With these benefits, the developed phototransistor array can work for the wide spectral image sensors and health monitoring (Fig. 3c). Subsequently, the group further developed the vertically stacked full-color QD phototransistor arrays for high-resolution photodetectors based on 3 nm CdSe QDs for blue-light detection, 5 nm CdSe QDs for green-light detection and 7 nm CdSe QDs for red-light detection (RGB detection).[93] The developed full-color flexible photodetector arrays can be integrated with a density of 5500 devices per cm^2, resulting in a photoresponsivity of 1.1×10^4 A W^{-1} and a strikingly high detectivity.

Moreover, QD photodetectors generally exhibit fast response speed even with the nanosecond fall time, enabling promising applications in fast monitoring. Sargent's group has contributed a lot to developing short-wave infrared QPDs with high response speed.[49,50,52] They put forward a resurfacing strategy for developing the enhanced coupling QDs with high hole mobility, which can enable a superior EQE of $\sim$70% and a specific detectivity over 10^{12} Jones at 1550 nm with a response time of 7 ns.[50] Such fast response speed may be attributed to the low exciton binding energy, high carrier mobility and favorable device structure, which can enable fast exciton dissociation, carrier transport and extraction for photodetectors even without bias.[94] In contrast, the high exciton binding energy and low carrier mobility may place great restrictions on the fast-response photodetectors.[95–97] Therefore, it is highly desirable to address the challenge *via* introducing versatile QD materials into organic photodetectors.

Additionally, QD materials also provided a powerful platform for developing the multi-role photodetectors. For instance, HgTe QDs can deliver the room-temperature photo-response beyond 5000 nm, which can cover the critical atmospheric mid-wavelength infrared window for remote sensing and night vision.[57] Guyot-Sionnest's group developed a two-terminal tandem QPD for dual-band infrared imaging based on the stacked HgTe QDs (Fig. 3d).[57] The tandem photodetectors can achieve the bias-switchable spectral response in two distinct bands centered at 2500 nm and 4000 nm, respectively (Fig. 3e). Accordingly, the developed dual-band QPDs exhibited a high detectivity of over 10^{10} Jones for dual-band infrared imaging (Fig. 3f). Additionally, Zhou *et al.* recently designed a facile QD-based upconversion photodetector, which can convert infrared photons from infrared light to visible light.[98] They employed PbS QDs as the infrared sensitive layer and CdSe/ZnS QDs as the visible emission layer for developing tandem upconversion photodetectors with high photon-to-photon conversion efficiency of 6.5% and a low turn-on voltage of 2.5 V. Moreover, the tandem photodetector can deliver a low J_{dark} of 1.3×10^{-8} A cm^{-2} and a high detectivity of 6×10^{12} Jones. With these benefits, the developed upconversion photodetectors exhibit great application potential in infrared detection and bioimaging. Nevertheless, most QPDs still suffer from high J_{dark}, which can even reach $\sim 10^{-5}$ A cm^{-2}, impeding the further improvement of detection performance.

Hybrid organic and QD photodetectors: future trends

From the above discussions, we can clearly see that OPDs have great detection performance at visible bands and the extremely low J_{dark}, while QPDs exhibit the broad detection range and fast response speed. Considering the strong complementarity of the two photodetectors, hybrid strategy holds the promising potential to be leveraged for developing the broadband and high-performance photodetectors. Organic semiconductors can significantly reduce the J_{dark} of QD photodetectors, which can further enhance the detectivity of QPDs. For instance, Wei *et al.* have demonstrated that the PBDB-T:PBI-Por blend can markedly suppress the dark current and enhance the photocurrent of QD photodetectors, resulting in a high responsivity of 6.32 A W^{-1} and a superior detectivity of 1.12×10^{13} Jones.[99] On the other hand, QD materials can also be employed for enhancing the detection performance of OPDs. Recent work by Han *et al.* has revealed that CdSe/CdS QDs and CdSe/ZnS QDs can work as the photomultiplication-inducing interlayer, which can trap the photogenerated electrons and induce hole injection from the electrode.[45] Using this mechanism, organic and QD hybrid photodetectors can achieve fast response speed, compared with that of conventional photomultiplication-type OPDs, which require a long time to induce carrier injection. Therefore, the hybrid photodetectors exhibited a high detectivity of 4.92×10^{12} Jones at 3 V bias and a fast response speed of 109 kHz, which was the record value for photomultiplication-type OPDs.

Additionally, organic and QD hybrid photodetectors can also be leveraged for multi-function applications. For instance, Yu *et al.* designed a novel upconversion photodetector with PbS QDs for infrared sensitive layer, which can offer electrons for organic light-emitting diodes (Fig. 4a).[100] With no bias and infrared illumination, the electron injection from the porous ITO electrode to the C_{60} channel is blocked, while under positive gate bias and infrared illumination, the generated electrons in PbS QDs can be transported to the ITO electrodes, therefore triggering the operation of light-emitting diodes (Fig. 4b). With these benefits, the developed upconversion photodetectors can achieve a striking EQE up to 100 000% and a high detectivity of 1.2×10^{13} Jones. Additionally, the hybrid strategy can also be employed for developing multispectral-sensing photodetectors. Pejović *et al.* recently designed a dual-band photodetector based on a tandem structure with an organic blend for visible and near-infrared response and PbS QDs for short-wave infrared detection (Fig. 4c).[101] The developed hybrid photodetectors exhibited a low J_{dark} of below 500 nA cm^{-2} (1 V), a high EQE of 70% in near-infrared bands and an EQE of 30% in short-wave infrared bands (Fig. 4d). Moreover, the photodetector can present the fast switch speed, with the fall time of 6 μs and 13 μs for OPD and QPD modes (Fig. 4e).

Summary and outlook

In this Highlight, we firstly discussed the features of OPDs and QPDs and revealed the great complementarity of these two

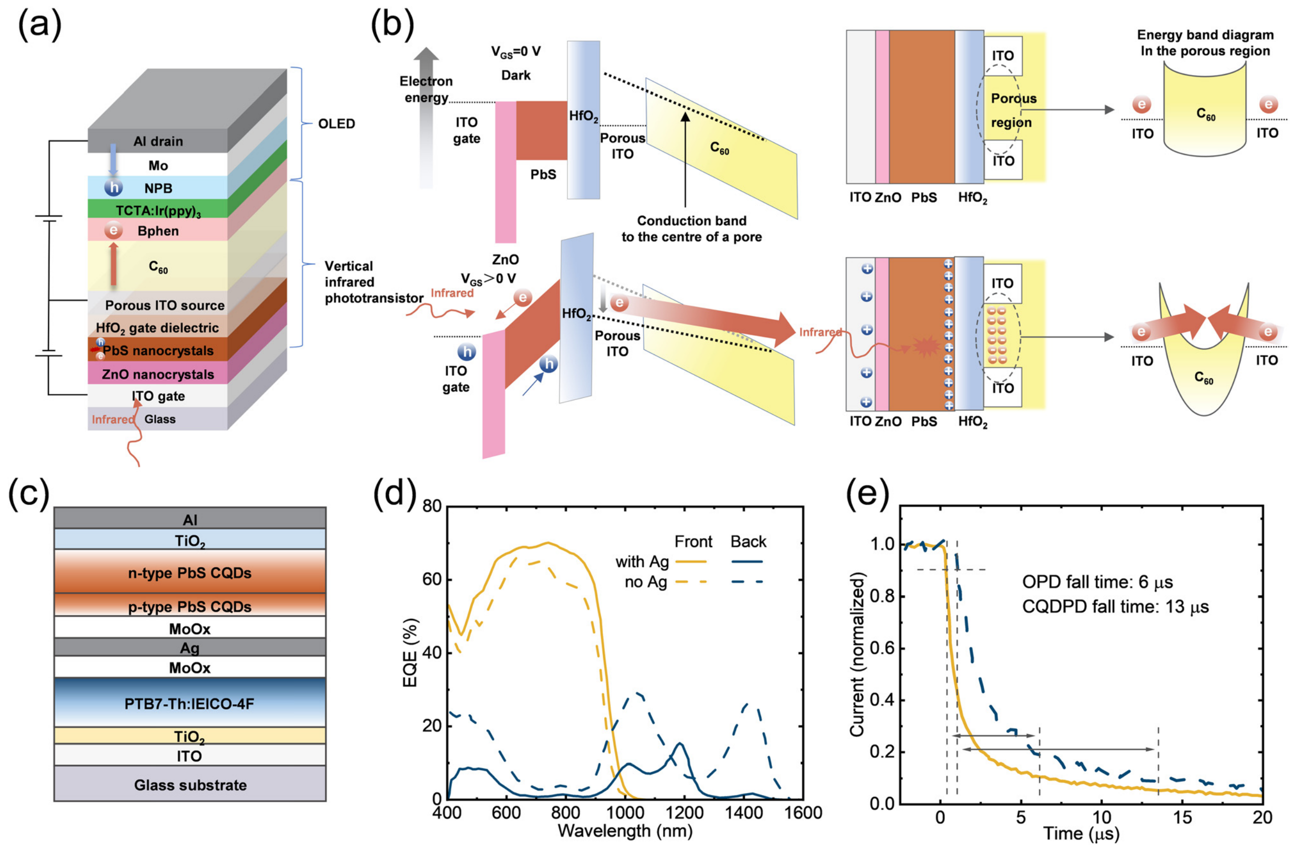

Fig. 4 The performance of organic and QD tandem photodetectors. (a) Schematic of the developed upconversion device. (b) Working mechanism of the upconversion device. (c) Schematic of the tandem device structure for dual-band detection. (d) External quantum efficiency of the developed tandem photodetectors. (e) The response speed of the developed OPDs and QPDs. (a and b) Reproduced with permission from ref. 100. Copyright 2016, Springer Nature. (c and e) Reproduced with permission from ref. 101. Copyright 2022, Wiley.

technologies. For OPDs, the superior flexibility, low dark current and great detection performance were highlighted and the slow response speed was also pointed out. For QPDs, broad detection, fast response and multi-function were underlined for their broad applications in image sensing, health monitoring, infrared communication and night vision. Additionally, we also discussed the recent progress of organic/QD hybrid photodetectors and highlighted the great promise of the hybrid strategy for developing perfect detection devices. Nevertheless, some tough challenges still placed great restrictions on the further advancement of hybrid photodetectors. For a bright future, the promising research directions of hybrid photodetectors are provided as follows:

(1) Enhancing response speed

Response speed is one of the critical parameters of high-performance photodetectors for fast detection applications. QPDs have been reported to deliver the superior response speed with the fall time of $\sim$10 ns, while OPDs generally have the long fall time of tens of microseconds. For photomultiplication-type OPDs, the fall time can even reach tens of milliseconds, which placed great restrictions on their commercial applications. Recent work by Saggar *et al.* revealed that the balanced electron and hole mobility is a critical benchmark for high-speed photodetectors and they found that tuning the blend ratio can significantly enhance the response speed from 0.8 to 4.5 MHz $f_{-3\text{dB}}$.[102] Nevertheless, the strategy is not applicable to photomultiplication-type photodetectors, which require long time to induce carrier injection and transport. Hybrid strategies based on organic and QD materials can offer a promising research direction for addressing this issue *via* bulk heterojunction or bilayer structures. QD materials with low exciton binding energy and high mobility can enhance exciton dissociation and carrier transport in the blend films, therefore resulting in fast photodetection.

(2) Reducing dark/noise current

Dark/noise current has a great influence on the detection accuracy of photodetectors. High dark/noise current significantly compromises the detection performance, especially for infrared QPDs in near-infrared, short-wave infrared to mid-wave infrared bands. Generally, the dark/noise current of QPDs was several orders of magnitude higher than that of OPDs at the same bias. Some promising reports have demonstrated that organic semiconductors can not only enhance carrier transport and extraction, but also markedly reduce the noise current of QPDs, highlighting the great potential of hybrid strategy on further improvement of detection performance.[45,99]

(3) Dual-band detection

Dual-band detection holds great potential in multi-function applications. For instance, dual-band detection in both visible and near-infrared bands can be employed to simultaneously achieve image sensing and health monitoring. Moreover, dual-band detection in both short-wave infrared and mid-wave infrared bands exhibits great promise in infrared imaging and remote temperature monitoring. Despite the great benefits, the existing dual-band photodetectors generally present the low response speed and detectivity, compromising their superiority in multi-role applications. Hybrid organic and QD strategies and the corresponding structure optimization may contribute a lot to address this challenge and further enhance the detection performance of dual-band photodetectors.

(4) Upconversion detection

Upconversion photodetectors have great applications in infrared detection and imaging. Such photodetectors can convert infrared photons to the visible lights, which can be directly recognized by the human eye and avoid the use of readout integrated circuits and display systems. QD materials can work as the infrared sensitive layer for detection and the hole/electron source, while light-emitting diodes can employ organic or QD materials for high-performance display. Nevertheless, the existing solution-based upconversion photodetectors can only convert near or short-wave infrared to visible lights mainly based on lead chalcogenide QDs. The direct display of mid-wave infrared lights based on HgTe QDs is highly desirable for remote temperature monitoring. More efforts should be devoted to this promising research directions.

(5) Flexible photodetectors

There exists the great need for flexible photodetectors in wearable electronics for image sensing and health monitoring. Nevertheless, QD photodetectors generally present the inferior mechanical performance, which places great restrictions on their applications in wearable electronics. The great flexibility of organic semiconductors[103] may offer promising roads to enhance the mechanical performance of QPDs *via* the hybrid strategy. Moreover, our recent work has demonstrated that a promising elastomer can further enhance the mechanical performance of organic semiconductors, which is favorable for developing stretchable electronics.[74] It can be expected that QD and organic hybrid photodetectors with a promising elastomer can simultaneously enhance the detection and mechanical performance, which can further broaden the application of this technology.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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