Emerging low-voltage organic phototransistors for high-gain photodetection and long-term memory

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Photodetectors enable sensitive conversion of optical signals into electrical signals, showcasing broad applications in biomedical imaging, environmental monitoring, optical communication, and underwater detection.[1-5] With the advancement of brain-inspired technologies, photodetectors are advancing toward memory-type devices that have capacities of both sensing and retaining optical information. They are desired to attain high gain and long-term information storage to meet growing demands of weak-light detection and data retention. Among them, high gain is crucial for realizing signal amplification and thus enabling low-cost readout circuits. Compared with inorganic counterparts, organic photodetectors (OPDs) possess advantages such as low cost, flexibility, and tunable spectral response, making them promising candidates for intelligent sensors and integrated sensing-memory-computing terminals.[2-5] Nevertheless, simultaneously achieving high gain and long-term information storage for OPDs under low-voltage operation remains challenging, either constrained by low gain or requiring the integration of multiple components to achieve information storage. Therefore, it is imperative to overcome limitations of conventional OPDs and develop novel organic permeable base phototransistors (Photo-OPBTs) with low-operating voltages featuring high gain and long-term memory characteristics (Figure 1).

Due to inherent properties of organic semiconductors, such as relatively low carrier mobility and high exciton binding energy, the gain mechanisms dominant in inorganic photodetectors are unsuitable for OPDs.[4] To address this issue, various gain strategies have been designed in photomultiplication-type OPDs (PM-OPDs) with a two-terminal vertical structure and phototransistor-type OPDs (PT-OPDs) with a three-terminal lateral structure, respectively.[5] Among them, PM-OPDs utilize carrier traps to capture photogenerated electrons or holes, assisting charge carriers with opposite polarity tunneling injection from the electrode under an external electric field, thereby enabling the photocurrent gain (Figure 1). Although the photocurrent of PM-OPDs rises efficiently than the dark current with increased bias voltages, the limit of PM-OPDs for weak-light detection is constrained by the dark and associated noise currents. Moreover, a relatively high bias voltage (≥ 15 V) is usually needed for PM-OPDs to achieve the optimum performance. For PT-OPDs, an electric field generated by the gate (G) bias facilitates dissociating photogenerated excitons into free holes and electrons. Then the photogenerated holes or electrons are trapped at the interface between a dielectric layer and an organic photosensitive semiconductor layer, while photogenerated carriers with opposite polarity are compelled away to form a lateral charge-transport channel, thereby achieving the photocurrent gain under an electric field between source (S) and drain (D) electrodes (Figure 1). However, PT-OPDs often suffer from drawbacks such as high-operating voltages, low responsivity, and slow response, which are closely related to the long lateral transport channel and the thick dielectric layer. The relatively slow response may limit PT-OPDs for application scenarios that require fast response with the time less than millisecond, such as fiber-optic communication, high-speed data transmission, and bioluminescence detection. Although the photogenerated carriers trapped at the interface can also be utilized for information storage, PT-OPDs still show relatively short memory retention time for the integrated sensing-memory function. It is still challenging for conventional OPDs to simultaneously afford high gain, low dark current, and long-term information storage under low-driving voltages (< 5 V).

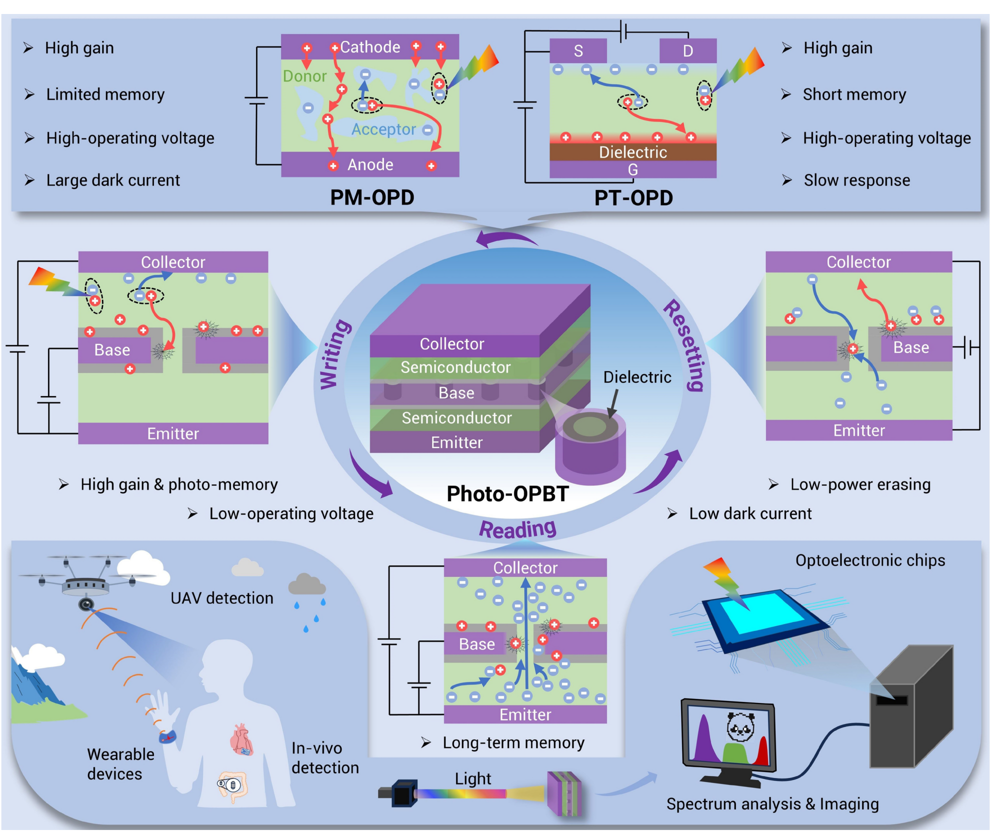

Figure 1. Gain and memory mechanisms as well as potential applications of Photo-OPBTs Top: working mechanisms of conventional OPDs. Middle: gain and memory mechanisms of Photo-OPBTs. Bottom: applications of high-gain, long-term memory Photo-OPBTs.

To solve this problem, Photo-OPBTs are emerging as a promising candidate.[2] Usually, this type of emerging OPDs consist of three electrodes (emitter, base, and collector) with at least two organic semiconductor layers to form a vertical structure (Figure 1). Numerous pinholes in the base serve as permeable transporting channels for carriers from the emitter to collector and function as potential barrier switches to suppress the dark and associated noise currents of devices.[3] After treated in the air, an ultrathin inorganic oxide insulating layer (~ 1-2 nm) forms both on the base surface and the walls of pinholes, which can significantly reduce leakage currents through the base. The vertical transport architecture of devices facilitates shortening carrier transport pathways, thereby enabling a strong internal electric field established within the channel even under a low-driving voltage to realize the high internal current gain. In addition, photogenerated carriers captured by traps at the interface between the organic photosensitive layer and the oxide dielectric layer generate a Coulomb electric field, which has two functions: (i) Assisting carriers injected from the emitter to permeate through the pinholes to the collector, thereby contributing to the photocurrent gain; (ii) Modulating the base potential of Photo-OPBTs for storing optical information.

Very recently, a low-driving voltage, high-gain, and long-term memory Photo-OPBT has been developed.[2] As illustrated in Figure 1, its integrated photo-memory function can be implemented through three working modes of writing, reading, and resetting, respectively. During the writing mode, photogenerated excitons in the photosensitive organic semiconductor layer will be dissociated into electrons and holes by an electric field between the collector and the base. Then, photogenerated holes will transport toward the base and be trapped at the interface, thereby storing the optical information and changing the base potential. During the reading mode, the Coulomb electric field, formed between trapped photogenerated holes and electrons injected from the emitter, is insufficient to switch the Photo-OPBT from the off state to the on state. However, the variation of the base potential can induce threshold voltages of Photo-OPBTs to shift gradually from positive to negative with the enhanced incident light. As a result, the Photo-OPBT achieves a high photocurrent gain to read out the stored optical information effectively under a low-driving voltage (≤ 2 V), with a high responsivity and a specific detectivity of up to 10^9 A/W and 10^{15} Jones, respectively. These representative quantitative metrics exceed corresponding average levels of ~10^6 A/W and ~10^{13} Jones for most conventional OPDs operated under higher voltages (≥ 5 V). Moreover, transfer characteristics of the Photo-OPBT under illumination reveal a pronounced hysteresis between forward and reverse sweeps, confirming an established optical memory window. Ultimately, the device maintains the stable read-out current for over 10^5 seconds, revealing the long-term memory capability of information storage. Although the Photo-OPBT exhibits stable repeating performance over 500 consecutive writing-reading-resetting cycles based on stably stored charges,[2] thermal detrapping of trapped photogenerated holes will induce the positive shift in threshold voltages for the base. To improve device stability, sealing the Photo-OPBT is also critical for shielding negative influences of environmental factors (e.g., oxygen and moisture) on internal organic semiconductors and associated photocurrents. Device simulations by the technology computer aided design (TCAD) further validate the photocurrent gain mechanism of the Photo-OPBT under low-voltage operation. During the resetting mode, a forward bias voltage (e.g., 2 V) is applied to the base while the collector maintains at 0 V. It compels the photogenerated holes remaining on the surface of the dielectric layer to be extracted by the collector or to recombine with electrons injected from the collector, thereby achieving the resetting process.

Compared with commercial devices and techniques, although Photo-OPBTs still face challenges such as stability issues and immature manufacturing processes, they show distinct advantages of natural flexibility, lightness, cost-effectiveness production, and integrated sensing-memory capacities without complex systems for signal denoising and enhancement. Obviously, the functions of Photo-OPBTs extend far beyond weak-light detection and imaging, showcasing great potential from common flexible wearable applications to emerging fields in optoelectronic chips and beyond (Figure 1). The long-term memory ability of Photo-OPBTs enables new possibilities for their utilization scenarios where real-time information transmission is impractical, such as unmanned aerial vehicle (UAV) detection operating in non-communicating states. Furthermore, the excellent flexibility and biocompatibility of Photo-OPBTs indicate their potential in wearable and/or implanted devices for high-gain detection and imaging, health monitoring, and in-vivo sensing. Additionally, the stored optical information can be demodulated by characteristic curves of Photo-OPBTs in the reading mode. This is crucial for identifying light at different wavelengths, making Photo-OPBTs and their integrated arrays suitable for spectrum analysis and imaging. More importantly, the low-power operation, fast response, high responsivity and long-term memory of Photo-OPBTs make them promising candidates for emerging applications such as logic gates, single-photon detection, artificial synapses, optoelectronic chips and soft circuits, thereby advancing the rapid development of semiconductor quantum computing and brain-like bionic intelligence.

In summary, the low-power Photo-OPBTs are emerging with integrated high-gain photodetection and long-term memory functions to surpass conventional PM-OPDs and PT-OPDs. Such Photo-OPBTs utilize traps on the dielectric surface to capture photogenerated carriers and store optical information, offering a novel approach for achieving ultrasensitive photodetectors with the integrated sensing-memory capability. Although Photo-OPBTs showcase good potential in photo-sensing, memory and imaging, advancing them from the laboratory to mature applications needs to address several challenges. These include the scalable fabrication of porous base electrodes, systematic exploration of advanced semiconductor/dielectric materials, and optimization of their carrier mobilities and interfacial trap densities to achieve higher detectivity and faster response, etc. The integration of artificial intelligence technologies such as data-driven algorithms and large models or agents into material screening, device optimization, and signal processing can efficiently accelerate the development of high-performance Photo-OPBTs and other emerging OPDs.

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AUTHOR CONTRIBUTIONS

Z. Y. conceptualized and supervised the study. P. D., G. R., J. W. and Z. Y. designed the manuscript. All authors contributed to the manuscript and approved the final version.

DECLARATION OF INTERESTS

The authors declare no competing interests.