Recent Progress on Advanced Optical Structures for Emerging Photovoltaics and Photodetectors

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Organic- and perovskite-based optoelectronics, which merge excellent optoelectronic properties with potentially large and high-throughput manufacturing, have attracted attention as emerging revolutionary technologies with considerable practical applications. Herein, the recent progresses in organic- and perovskite-based photovoltaics and photodetectors with integration of judicious optical structure designs are summarized. The characterization and performance metrics of such devices from the perspectives of device architecture, physics, and material science are studied. Research related to devices having dielectric mirrors, diffracted Bragg reflectors/photonic crystals, microcavities, and 2D photonic structures as design elements is discussed. Some suggestions of promising directions for future studies are concluded.

1. Introduction

The continued integration of optoelectronic technologies for energy consumption, environmental monitoring, machine vision, automated unmanned driving and factories, image processing, and the Internet of things relies on new developments in photovoltaics (PVs), light sensors, light-emitting diodes, and lasers. Because of the myriad attractions of organic and perovskite materials—including their manipulable light absorption,[1,2] flexibility, light weight, and potential for high-throughput manufacture[3] —and their successful application in PVs, these materials are becoming important components in other optoelectronic applications. The power conversion efficiencies (PCEs) of perovskite solar cells (PSCs) and organic photovoltaics (OPVs) have skyrocketed in the past 10 years to over 25% and 18%, respectively, further hastening their applicability. Park and Zhu provided a review of solution/vapor deposition techniques for the fabrication of large-area PSCs and of technologies for achieving high-performance modules.[4] Other review articles have discussed the fundamental properties of these materials and their applications. Xu and Lin reported the recent advances in photodetectors (PDs) based on these emerging materials.[5] The Rahmany and Etgar and Shi et al. independently described progress in semitransparent PSCs from both materials and device perspectives.[6] Hu and coworkers highlighted the state-of-the-art protocols for constructing qualified infrared (IR) organic photodetectors (OPDs) for integrated applications.[7] In this Mini Review, we focus on recent advances in organic and perovskite materials that integrate with optical design, especially for their applications in PVs and PDs.

Approaches for optical manipulation in PV applications can be divided mainly into spectral control and optical design. By controlling the bandgaps of various conjugated molecules or perovskites, it is possible to fabricate devices having vivid colors, or that are highly transparent, for building-integrated PV applications.5,8,9 Using thin metallic electrodes (<20 nm) and active layers that mainly harvest ultraviolet (UV) and near-infrared (NIR) light has led to high-performance semitransparent PVs (ST-PVs).[10–17] The integration of new structures with low-bandgap active layer materials has provided high-performance visibly transparent OPVs.[18–24] For instance, Yang et al. demonstrated an ST-OPV having a PCE of 12% and an average visible transmittance (AVT) of 20% using a thin Au/Ag electrode and a strategy of transparent hole-transporting frameworks.[25] Several light-trapping methods, including the incorporation of antireflection layers,[26,27] microcavity (MC) structures,[28] distributed Bragg reflectors (DBRs) and photonic crystals (PCs),[29,30] and nanostructures,[31,32] have further optimized the light harvesting and optical responses of such devices. Shen and coworkers reviewed the use of MCs in OPVs[28] whereas Lova et al. highlighted the use of solution-processed...
planar 1D PCs (organic material- and inorganic material-based DBRs) with considerable application interest. Figure 1 shows many of the recent advances in OPVs and PSCs. As a selected example, Xie et al. demonstrated a ST-SC device featuring MoO$_3$/Ag/WO$_3$ as the transparent top electrode and displaying a PCE of 15.33% and an AVT of 12.18%. Liu et al. used an oblique incidence decoupling technology to obtain a light use efficiency (LUE) of 4.83%—at the time, the record for an OPV. This record was overcome by Forrest and coworkers, who demonstrated a ST-OPV having a PCE of 10.8% and a visible transparency of $\approx$50%.

With their ability to convert incident light to output electrical signals, PDs are indispensable devices that are used widely, for example, in the charge coupled devices of cameras, in the receivers for remote control of appliances, and in optical communications. Unlike PSC and OPV devices that are operated under positive bias for power generation, PDs are operated under negative bias voltages. When a sufficient reverse bias is applied, the induced photogenerated charges are extracted at corresponding electrodes; notably, the photocurrent generated under operation is independent on the applied voltage. There are many measurable characteristics for evaluating the performance metrics of PDs, including the external quantum efficiency (EQE, the ratio of incident photons converted to electrons), the spectral responsivity ($R$, in units of A W$^{-1}$), the frequency response and response time (the time required to collect generated charge carriers), the noise equivalent power (NEP, the minimum detectable incident light power), the specific detectivity ($D^*$, in units of cm Hz$^{1/2}$ W$^{-1}$), the photocurrent and dark current density ($J_0$, the current generated under dark at reverse bias), and the linear dynamic range (LDR, the linear response range of the photocurrent upon varying the incident optical power). Unlike standardized measurement and calibration method are applied to direct compare the performance of OPVs and PSCs. The reported metrics of PDs are often obtained under different experimental conditions (e.g., various applied bias voltages or light intensities) for different applications, making it difficult to directly compare the results of an organic or organic hybrid PD with those in the literature. In this Review, for the most important figures of merit of PDs, we highlight the specific optical designs, applications, and main experimental methods, along with their pros and cons, to provide a better understanding of the progress of these optoelectronic devices. In comparison with the absorption coefficients of silicon ($\approx$10$^5$ cm$^{-1}$), those of organic semiconductors and perovskites are much higher ($\approx$10$^7$ cm$^{-1}$), enabling them to absorb a sufficient number of photons in a thin active layer (up to several hundreds of nanometers). Because of their low permittivities ($\varepsilon_r \approx$3–4), organic semiconductors have high exciton binding energies; therefore, OPDs have mainly featured a bulk heterojunction (BHJ) active layer, prepared by blending electron donating (D) and electron accepting (A) moieties, for efficient exciton dissociation and free carrier transport. Perovskite materials are useful because of the synergistic effect of both inorganic and organic semiconductors (e.g., compatibility with solution- and vacuum-deposition and their related processing temperatures). For example, Lin et al. demonstrated a broadband perovskite PD (PePD) having low noise and fast temporal response. The incorporation of tin cations in lead perovskites has led to devices displaying efficient NIR responses. Variations in composition tailored the responsivity of the PePD, whereas variations in the molecular structure and judicious selection of the D/A moieties led to OPDs covering different response wavelengths.

We begin this Review by highlighting the state-of-the-art protocols for constructing ST-OPVs and -PSCs with focus on recent emerging concepts and advances in these materials integrated with various photonic structures (Table 1). We then discuss recent advances in PDs for integrated applications. We are aiming to provide the reader with a comprehensive understanding of optical designs for future integrated practices of organic- and perovskite-based PVs and PDs. For the most relevant metrics of these optoelectronics devices, we discuss a variety of optical methods for obtaining them and examine their dependence on the experimental conditions. We conclude with a discussion of the specific applications, challenges, and limitations of these devices—hopefully to increase awareness and facilitate a more critical understanding.

2. Principles of Photonic Design

DBRs, also known as periodic 1D PCs or dielectric mirrors (DMs), have been used to selectively reflect incident light for reharvesting in PV devices. As shown in Table 1b, DBRs have multilayer structures with alternating layers of high ($n_1$) and low ($n_2$) refractive index. The relationship between the different layers within a DBR structure can be expressed by the following equation

$$n_1 \times d_1 = n_2 \times d_2 = \frac{\lambda_0}{4}$$  \hspace{1cm} (1)

where $d_1$ and $d_2$ are the thicknesses of the layers of the PC and $\lambda_0$ is the reflection center wavelength. In a DBR, the optical path is equal to a quarter of the value of $\lambda_0$, such that the reflected light from every interface has an identical phase, leading to
constructive interference. In most OPVs, using a DBR will increase the degree of light harvesting that is lost by having a transparent electrode (i.e., without reflection from the top electrode). Because of the periodic variations in the refractive indices in DBRs or PCs, these devices can feature various photonic bandgaps in one, two, or three dimensions (Table 1c). The unique properties of PCs that allow the manipulation of the wavelength of light and control the directions in which it can travel are useful in optoelectronics.

The typical MC structure used in OPV applications is the Fabry–Pérot cavity (FP cavity) (Table 1d), also known as a planar parallel cavity, which has one dimension of size that is on the order of magnitude of the wavelength of light. By incorporating two parallel planar metal films (i.e., mirrors of high reflectivity) and a layer sandwiched between them, light within the MC structure reflects, resonates, and oscillates between the “mirrors” and leads to controllable resonance within the device. Many theoretical and experimental studies and physical models of FP resonant cavities have been conducted, the details of which can be found in earlier reports. Consequently, various resonance modes can be realized by controlling the thickness, composition, and cross-section of metal films with MC structures.

### 3. Optical Assessment

The use of an optical assessment structure, as shown in Table 1, can improve the performance of an opaque PV. For instance, by optimizing the in-cell spacer layer (MoO₃) and out-of-cell capping layer (MoO₃ or LiF), Lin et al. prepared a highly efficient top-illuminated small-molecule-based indium tin oxide (ITO)–free-opaque OPV having the structure Ag/spacer layer (MoO₃)/active layer/Ag/capping layer. The use of a TeO₂/MoO₃ composite layer that induced an MC effect within the active layer increased the performance of opaque PV devices. In these cases, incident light was reflected and efficiently captured within the MC structure, thereby amplifying the electric field and improving the PCE. In this Review, we focus on such device technologies that can lead to improved performance.
provide applications with semitransparency or vivid colors, according to the penetration or reflection of light. Table 2 shows the parameters and abbreviations used for defining these devices. Herein, the color purity and transparency of the devices are determined according to the CIE 1931 standard colorimetric system. The corresponding color temperature (CCT) is the color temperature (Planckian locus) of the light source located in the CIE 1931 color space. The color rendering index (CRI) is defined as the degree of change between the transmitted and incident light. A high CRI represents high neutral chromaticity. About 53% of the photons in the solar spectrum have wavelengths in the range 780–2500 nm, of which the NIR photons can be used for photovoltaic conversion and the IR photons can be reflected to achieve heat dissipation. Infrared photon rejection rate (IRR) is the effective reflectivity of IR photons, and the higher value showing better the heat dissipation effect.

### 4. Semitransparent OPVs and PSCs

#### 4.1. Dielectric/Metal/Dielectric Electrodes

To realize semitransparent OPVs and PSCs, one can manipulate the coverage, thickness, or bandgap of the active layer or replace the opaque metal electrode with light-transmitting media (e.g., metal nanowires, transparent conducting oxides, transparent conducting polymers, graphene, and carbon nanotube). Although the most convenient way to fabricate a ST-PV is to decrease the thickness of the top metal electrode and, thereby, increase its transparency, there is always a trade-off between conductivity and transparency. By sandwiching a thin metal film between dielectric layers, decent electrical and optical properties can be achieved.

DIELECTRIC/METAL/DIELECTRIC (DMD) electrodes can optimize the performance of PVs in terms of their transparency, conductivity, and mechanical flexibility, further the inducing of microcavity can enhance the performance of the device. In OPV study, Sano et al. used a MoO3/Ag/MoO3 MC electrode to prepare a neutral color ST-OPV, controlling the spectral distribution of transmitted light through fine-tuning of the blend ratio of the active layer and the thickness of the Ag layer (i.e., 10 nm). They reported a ST-OPV having a PCE of 4.0%, an AVT of 39%, and a CRI of 97 (Figure 2a). Sun et al. demonstrated a PFBDB-T:C8-ITIC-derived ST-OPV having the same MoO3/Ag/MoO3 electrode; it exhibited a PCE of 9.2% and an AVT of 22%. Sun and coworkers studied all-NIR-absorbing materials having the same DMD structure (Figure 2b). They used two conjugated molecules, PDTTP-DFTB and FOIC, to harvest solar light efficiently in the range of 500–1000 nm. The thickness of the top MoO3 layer was varied for optical management to alter the transmittance of the device and optimize the VLT. The optimized devices provided excellent VLTs of 52–61.5%, PCEs of 3.5–4.2%, good CRIs (>90), and color rendering properties. Lim et al. used emissive 22 Au quantum clusters as an interfacial layer between the active layer and the MoO3/Ag/MoO3 electrode (Figure 2c). In comparison with the normal device, the device incorporating the 22 Au quantum clusters (Au22-QCs) exhibited improved performance under low-intensity sunlight and greater flexibility. The DMD structures in ST-PSCs have been similar to those in ST-OPVs. For example, Hanmandlu et al. introduced a DBR (LiF/MoO3) structure to improve the VLT (Figure 2d). In addition to providing a high dielectric constant, the BCP also functioned as a diffusion buffer layer between the electron transport layer (ETL) and the Ag electrode. The optimized device demonstrated a PCE of 13.49% and an AVT of 11.3%. Della Gaspera et al. incorporated a MoO3/Au/MoO3 top electrode in their ST-OPV (Figure 2e). Their best device, which featured an active layer of methylammonium lead iodide (MAPbI3) that was >55 nm thick, gave a PCE of 5.3% and AVTs as high as 31%. Xie et al. reported an ST-OPV having a MoO3/Ag/WoO3 electrode, with the WoO3 enhancing the transmittance in the long wavelength region (Figure 2f). Their best device featured FA0.5MA0.5Cs0.12PbI2.06Br0.96 as the active layer and provided a PCE of 13.16% and an AVT of 16.55%.

#### 4.2. DBRs

A DBR or PC structure can effectively change the reflection spectrum of an OPV. Zhang et al. used a three-layer LiF/MoO3 DBR to prepare a high-performance (PCE = 9.37%, AVT = 20%) ST-OPV having a CRI approaching 100. In 2018, Sun et al. introduced a DBR (LiF/MoO3) structure to improve the VLT selectively while maintaining high reflectivity toward NIR light. They obtained an ST-OPV displaying a PCE of 6%, a VLT of 25%, and an IRR of 80%. Efficient blocking of IR radiation endowed their ST-OPV with excellent heat-insulation properties meeting commercial requirements. Through precise design of a DBR layer (TeO2/LiF/TeO2/LiF), Li et al. improved the VLT to 30%, with a PCE of 9.3% and an IRR exceeding 93% (780–2500 nm). Chen et al. embedded an additional optical adjustment layer of P3HT:ICBA under the WO3/LiF structure to adjust the phase of the incident light, increase light harvesting, and maintain the CRI. Xia et al. examined the effect of the number of DBR layers and the thickness of the LiF/MoO3 structure through high-throughput optical engineering on multi-objective (VLT-PCE) optimizations. They demonstrated that changing the DBR structure could optimize the generated photocurrent while maximizing the VLT (PCEs of 7.5–9% and VLTs from 40% to 20% could be expected). Very recently, Wang et al. positioned a LiF/MoO3 DBR structure on top of an ITO/poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS)/PBDB-T:F6:BTTPC/PFN-Br/Ag device...
They used new ternary blends with an alloy-like NIR acceptor and photonic reflectors, together with optical simulation, and effectively improved the PCE while maintaining the AVT. This excellent performance led to a PCE of over 12%, an optimized IRR of 90% (780–2500 nm), and an AVT of 23%. Their simple DBR-integrated ST-OPV combined the best PV performance with heat dissipation. Liu et al. reported largely decoupled light transmission from incident light when using an IR-shifted nonfullerene acceptor (NFA)-derived device. They prepared 1D PC/DBR-based devices having three-layer (LiF/MoO3/LiF) and five-layer (LiF/MoO3/LiF/MoO3/LiF) structures and studied their performance over a wide range of incident angles. They obtained a PCE of 9.67% at 50° incidence and an AVT of greater than 50% at normal incidence. Zheng et al. used a polymer 1D PC structure with a co-extrusion of poly(methyl methacrylate)/polystyrene (PS) stacks (3M). They integrated one to three layers of the polymer 1D PC on the polyethylene terephthalate (PET) side and obtained flexible ST-OPVs having the
The wide distribution of CIE color coordinates for their devices suggests that such flexible ST-OPVs may be suitability for arc-shaped building and portable devices. Ravishankar et al. applied a PC (LiF/MoO$_3$/LiF/MoO$_3$) to adjust the spectral absorption of ST-OPVs to minimize the attenuation of sunlight and achieve net-zero-energy greenhouses (Figure 3b). They developed an energy model to simulate the energy load, solar power generation, and light entering the greenhouse.

4.3. DBRs with DMD Electrodes

To achieve highly vivid color transparency, researchers have tended to use hybrids of MC electrodes and DBRs. In 2018, Zhong et al. used WO$_3$/Ag/WO$_3$ MC electrodes (anodes) to improve the translucent optical performance. Further integration with a PC structure having eight pairs of WO$_3$ multi-layers increased the performance of the ST-OPV (PCE = 8.37%; VLT = 21.7%). The combination of MC and PC structures could
also tune in a broad range of see-through colors by tailoring the center wavelength of the photonic bandgap. The use of an outcoupling layer can further optimize the optoelectronic properties of ST-OPVs. Li et al. adopted an outcoupling layer of 4,4'-bis(N-carbazolyl)-1,1'-biphenyl (CBP)/MgF$_2$/CBP/MgF$_2$ with a multilayer SiO$_2$/SiNx DBR and an antirefection coating to improve the degree of light manipulation (Figure 3c).$^{[83]}$ They obtained a maximum LUE of 3.56 ± 0.11%, a PCE of 8.0 ± 0.2%, CIE coordinates of (0.337, 0.349), and a CRI of 87. Very recently, Forrest and coworkers prepared an ST-OPV providing a PCE of 10.8% and visible transparency of $\approx$50%.$^{[111]}$ By combining an NIR-harvesting material, an antirefection coating (ARC; SiO$_2$/MgF$_2$ on the ITO side), an optical outcoupling structure (MgF$_2$/CBP/MgF$_2$/CBP on top of the electrode), and a transparent electrode (MoO$_3$/Ag), they overcame the trade-off between PCE and transparency.

5. Colorful OPVs and PSCs

5.1. Photonic Structures That Alter the Transmission Spectra of ST-PVs

The perceived color of a solar cell can be modified by changing its reflection or transmission spectrum. To change the transmission spectrum of a ST-PV, MC electrodes and a DBR can be introduced on the back side of a solar cell, thereby allowing the passage of light in only a specific spectral range.

5.1.1. Metal/Dielectric/Metal Electrodes

By tailoring the thickness of the dielectric layer, the penetration of specific wavelengths can be controlled to achieve high-purity color appearance. Lin and coworkers used a Ag/N,N'-di(1-naphthyl)-N,N'-diphenyl-(1,1'-biphenyl)-4,4'-diamine (NPB)/Ag MC to adjust the color of a small-molecule-based ST-OPV.$^{[144]}$ They obtained excellent color-control capability by varying the thickness of the NPB (optical spacing) layer; the maximum transmission peak ($T_{MAX}$) of the devices could be tuned from blue to red with a full width at half maximum (FWHM) of 30 nm (Figure 4a). Notably, most of the dielectric layers were metal oxides; their findings demonstrate a good example of a small-molecule-based MC for OPV applications. Chen and coworkers developed a vivid color ST-OPV by embedding a Ag/ITO/Ag MC structure.$^{[145]}$ They obtained the wide color gamut by precisely controlling the thickness of the ITO layer. The PCEs of their green, yellow-green, and yellow PTB7-Th:PC$_{71}$BM-derived ST-OPVs were greater than 8%, with values of $T_{MAX}$ of greater than 14.5%. In a previous study, they had used the same MC.

Figure 4. a) Device architecture of MC-embedded color-tunable transparent OSCs. b) Schematic representation of hybrid-electrode-mirror MC-based ST-OSCs with PTB7-Th:PC$_{71}$BM as the active layer. c) Device structure of color filter (CF) integrated OPVs and photograph of semitransparent reference, blue, green, orange, and red CF-OPVs. d) Schematic representation, simulated (dashed lines) and measured (solid lines) spectral transmittance curves, and photographs of colorful ST-PSCs incorporating Ag/ITO/Ag MCs. a) reproduced with permission.$^{[144]}$ Copyright 2013, Wiley-VCH. b) Reproduced with permission.$^{[147]}$ Copyright 2019, American Chemical Society. c) Reproduced with permission.$^{[148]}$ Copyright 2019, American Chemical Society. d) Reproduced with permission.$^{[149]}$ Copyright 2016, American Chemical Society.
structure in NIR-based binary and ternary active layers to alter the color of an ST-OPV.[84] Kim et al. applied a Ag/TiO$_2$/Ag metal/dielectric/metal (MDM) electrode to obtain PTB7-Th:PC$_{71}$BM ST-OPVs having values of $T_{\text{MAX}}$ of greater than 25% and PCEs of up to 4.5%. [146] Zhong et al. embedded ultrathin Au as an interlayer between the active layer and the Ag/VO$_2$/Ag structure, resulting in PCEs of up to 9% and values of $T_{\text{MAX}}$ in the range 10–25% for various color gamut samples (Figure 4b).[147] Shafian et al. applied solution-processed TiO$_2$-AcAc as the dielectric layer (Figure 4c).[148] wet-processing of the dielectric layer contributed to the facile fabrication of their ST-OPVs.

For PSCs, Lu et al.[149] and Lee et al.[150] used MC electrodes, comprising a dielectric layer (ITO and WO$_3$, respectively) sandwiched between two thin Ag layers, on the back side of PSCs. As shown in Figure 4d, by carefully altering the thickness of the dielectric layers within the MC structures, Lu et al. could broadly tune the transmission color of their ST-PSCs from blue to reddish-orange; their yellow device exhibited the highest PCE of 7.1% while maintaining promising long-term stability (for 70 days under Ar in the dark).[149] Lee et al. introduced ultrathin 3,4,9,10-perylenetetracarboxylic bis-benzimidazole (PTCBI) layers preceding each Ag layer to form smooth Ag surfaces in the MC structures and minimize the light scattering loss; they demonstrated red, green, and blue ST-PSCs with PCEs of $\approx$4%.

Lee et al. exploited a more complicated MC electrode in a Ag/SiO$_2$/ZnS/Ag/ZnS configuration to demonstrate red, green, and blue ST-PSCs having PCEs of 10.47%, 10.66%, and 11.18%, respectively.[153] The ZnS as the middle cavity medium determined the transmissive colors, while the SiO$_2$ between the bottom Ag electrode and the ZnS cavity prevented direct contact between the conductive ZnS and the underlying BCP layers. The top ZnS layer served as a phase-compensating functional layer, providing not only an additional reflection phase shift to further improve the incident-angle-dependent performance but also better impedance matching to enhance the transmission efficiency.

5.1.2. DBRs

Shen et al. embedded Ag/Au nanoparticles (NPs) in OPVs and integrated them with WO$_3$/LiF to induce local surface plasmon resonance (SPR) effects to improve the optoelectronic properties of ST-OPVs (Figure 5a).[125] By adjusting the center wavelength of the PC, they obtained a CRI of 95, a CCT of 5340 K, an AVT of 20.38%, and a PCE of 7.07 ± 0.16%. Subsequently, they used the same PC structure and embedded an additional layer of WO$_3$ in ITO/PFN/PTB7-Th:PC$_{71}$BM/MoO$_3$/Ag/VO$_2$/LiF to gradually red shift and broaden the spectrum of the device (Figure 5b). They obtained devices having PCEs of...
9.36 ± 0.16% and an AVT of 14.31 ± 0.65% with controllable color.[85] To avoid the trade-off between the transparency and color purity (the narrow bandpass of transmitted light) of an ST-PV, Wang et al. developed a spectrally selective electrode (SSE, Al:Ag/ LiF/NPB/carbon fluoride) that allowed the efficient transmission of selected light with a narrow bandwidth (Figure 5c).[126] They obtained color ST-OPVs with PCEs of up to 15.07%, with color purity close to 100% and values of $T_{MAX}$ close to 30%.

5.2. Photonic Structures That Alter the Reflection Spectra of Solar Cells

Another way to give color to a solar cell is to change its reflection spectrum, either by changing the device structure and, thus, the optical interference in the device or by incorporating a 1D or 2D photonic structure. Huang et al. incorporated self-assembled organic molecules into a nanostructured cover layer to create a gradient index film that effectively increased the amount of light entering the device, increasing the PCE by 44%.[133] Dudem et al. used a broadband/omnidirectional subwavelength PC architecture as the AR layer of such a device, affecting the light scattering and increasing the light harvesting.[134] Their structure was fabricated on the surface of a polydimethylsiloxane (PDMS) layer through soft imprinting lithography (SIL) using anodic aluminum oxide (AAO) molds (Figure 6a). When compared with the normal device, the PCE (from 8.3% to 10.2%), the AVT (from 23% to 26%), and the CRI (from 84 to 90%) all improved. The Khezripour group incorporated a double-sided TiO$_2$ nanoarray grating structure to increase the performance of...
their OPV.[135] Lan et al. used nanoimprinting to prepare a 2D photonic structure that enhanced omnidirectional and broadband light harvesting (Figure 6b).[136] Zheng et al. proposed a similar method.[137] In ST-PSC research, Wang et al.[138] Li et al.[139] and Jiang et al.[140] changed the perceived color of their ST-PSCs by adjusting the thickness of the rear transparent electrodes, thereby varying the optical interference within their devices. Wang et al. and Li et al. used sputtered ITO and Jiang et al. used conductive PEDOT:PSS (prepared by transfer printing) as their rear transparent electrodes. The PSCs prepared by Jiang et al. exhibited PCEs of 12.8–15.1% and 11.6–13.8% (from red to blue) when illuminated from the FTO glass side and from the rear PEDOT:PSS electrode, respectively.

Kim et al. prepared inverted moth-eye-structured PDMS through soft lithography and attached it to the glass substrates of their PSCs.[138] When the lateral pitch of the moth-eye structure was 1000 nm, the structured PDMS acted as a reflection grating and gave the PSC an iridescent appearance, with a PCE of 17.43%. As shown in Figure 5d, Yoo et al. used alternating high-index TiO₂/low-index SiO₂ multilayer structures to create narrow-bandwidth reflective filters on the substrate, obtaining red, green, and blue PSCs with PCEs of up to 18.9%.[142] Compared with typical DBRs that feature periodic structures, the nonperiodic structures they exploited eliminated the baseline ripple-shaped patterns in the reflectance spectra (arising from the secondary constructive interference and widening the color gamut). The shape of the reflectance spectra of the reflective filters became more rectangular as the number of stacked layers increased, whereas the peak wavelength of the reflective filters moved to longer wavelength as the thickness of each multilayer increased. Lee et al. used nanoimprint lithography to deposit Ag nanostrips at the subwavelength scale on the outer surface of the glass substrate (Figure 6c).[138] The subwavelength Ag nanostrips displayed localized SPR and a structural reflective color for transverse magnetic (TM)-polarized light that could be altered by changing the width of the nanostrips. The fabricated red, green, and blue PSCs exhibited PCEs of 10.12%, 8.17%, and 7.12%, respectively. In addition to positioning them on the front surfaces of PSCs, photonic structures could also be embedded in the PSCs to alter the reflective spectra of the devices. Zhang et al. created a PC scaffold comprising alternating dense TiO₂ and porous SiO₂ layers; the porosity of each layer was varied to maximize the refractive index contrast between the layers in the photonic structure.[129] To prevent infiltration of the TiO₂ precursor into the porous SiO₂ layer during spin coating, the SiO₂ NP solution was mixed with a polymer that could be removed through subsequent high-temperature sintering. By modifying the thickness of the individual layers, the reflective spectra of the PSCs could be tuned from red to blue, with the blue PSCs exhibiting the highest PCEs (up to 8.8%). When compared with the addition of reflective filters on the outer surfaces of PSCs, integrating the photonic structures within the PSCs dramatically minimized the angular variability of the perceived color. Wang et al.[139] and Liu et al.[140] created nanostructured ETLs by removing the PS microspheres after depositing the ETL. The colorful PSCs fabricated by Liu et al. used an inverse opal-structured SnO₂ and SnO₂–TiO₂ composite and delivered PCEs of up to 16.8%; those made by Wang et al. featured TiO₂ nanobowl arrays and exhibited PCEs of up to 16.94%.

The periodic morphology of the TiO₂ nanobowl arrays was preserved after deposition of the perovskite active layer, the hole-transporting material, and the top electrode, resulting in the fabricated PSCs exhibiting angle-dependent structural color.

In addition to the strategies mentioned earlier, the perovskite itself can also be engineered to create photonic nanostructures within a device. Deng et al. used doctor-blade coating to deposit perovskites having concentric ring photonic structures with near-equal ring spacing in each perovskite domain during solvent evaporation (Figure 6d); this phenomenon can be explained by alternating pinning and depinning processes at the edge of the solution droplet.[141] By tuning the concentration of the precursor solution and the temperature during doctor-blade coating, they could control the domain size and ring spacing and, thus, the reflective colors of the fabricated PSCs when observed from the metal top electrode. PSCs based on these nanostructured perovskites exhibited a stabilized PCE of 12.2% and an illumination angle-insensitive (from 10° to 50°) short-circuit current density (JSC). Meng et al. removed the monolayer of closed-packed PS microspheres after they had been infiltrated with spin-coated perovskites, resulting in 2D inverse-opal-structured perovskite thin films.[142] The fabricated PSCs exhibited PCEs of up to 11.2% and vivid colors that varied depending on the viewing angle and the diameter of the templating PS microspheres. Wang et al. used nanoimprinting to introduce large-area gratings into the perovskite active layers, using commercially available CD-R and DVD-R disc recordable discs as initial masters.[143] The periodic ordered micronanostructures on the perovskites not only induced a colorful appearance but also enhanced the degree of light trapping in the solar cells. These colorful PSCs delivered exceptionally high PCEs of up to 19.71%—the highest achieved to date for colorful PSCs.

In addition to the aforementioned optical designs, luminescent solar concentrators (LSCs) have also been recognized as efficient light-management media and LSC-integrated PVs have been spotlighted as building-integrated photovoltaics (BIPVs) providing high transparency and an alternative optical design for OPVs and PSCs. In an LSC panel, luminescent materials absorb the incoming light and re-emits the energy in the form of light that is subsequently waveguided to the edge PV and converted to electricity. Various LSCs have been demonstrated, such as the polymeric[196] or inorganic host systems.[197] The embedding of quantum dots (QDs),[198] organic dyes,[199] carbon dots,[200] and bulk CH₃NH₃PbBr₃ perovskite fluorophores[201] in above hosts system were elucidated thorough their optoelectronic and PV characterization. In 2015, Chen and coworkers embedded the organic dyes in PDMS as their LSC and induced a strong scattering effect by controlling the segregation of the dyes. Their LSC effectively waveguides the light toward the Si-based PV and leads to a record performance of the devices.[202] Li et al. studied the impacts of the luminophore concentration and the roughness of waveguide on the PCEs of their devices.[203] They observed an improvement of the photostability of printed OPV devices. Hong and coworkers coupled the LSC with OPV for harvesting indoor and outdoor light.[204] In their study, they observed an optical efficiency and a PCE of 8.5% and 0.17%, respectively, under 1-sun illumination. Moon and coworkers demonstrated a combination of the up-conversion-assisted dual band-harvesting LSC
for PSC which exhibited a high AVT of 82% and a maximum PCE of 7.53%.[205]

6. Advanced Optical Design for PDs

In general, PDs can be divided in terms of their geometries into two-terminal PDs and photoconductors and three-terminal phototransistors. Herein, we discuss the two-terminal PD structures having geometries similar to those of OPVs and PSCs. Again, the quality of a PD is closely associated with its molecular design (i.e., its active layer) as well as the configuration of the device. Innovative optical design can not only optimize the device performance but also realize specific applications without any change to the active layer. In this section, we discuss recent progress and excellent examples of advanced optical engineering for high-performance OPDs and PePDs.

When using a metal–metal resonant MC for PD applications, the mirrors on both sides of the active layer simultaneously serve as electrodes, while the detection wavelengths can be modified by varying the cavity thickness (the active layer, the transporting layer, or both). Tang et al. used a resonant optical cavity (bottom electrode: 30 nm Au and top electrode: 100 nm Ag) device structure for wavelength-tunable light detection (Figure 7a).[130] Their device was highly wavelength-selective by adjusting the thickness of the active layer (MC effect); they obtained an NIR PD operating with high resolution (down to 14 nm), and low dark currents. Furthermore, using a PEDOT:PSS–derived DBR to replace the Au layer led to a superior specific detectivity (up to $10^{13}$ Jones). Furthermore, their findings have broadened the available selection of active layers by exploiting the intermolecular charge transfer (CT) absorption band between the intercalated donor and acceptor. The CT effect extended the detection wavelengths of PD beyond those of the neat absorber. Mischok et al. used high-reflectivity DBR (10 $\times$ TiO$_2$, SiO$_2$) pairs to prepare an OPD having an EQE peak at 900 nm with an FWHM of 22 nm (Figure 7b).[131] Xu et al. integrated a metasurface with broadband functionality into an MC having the structure Si/SiO$_2$/metasurface Au/ZnO/active layer/Au (Figure 7c).[132] They observed large gains in responsivity from 1.5 $\times$ to 2 $\times$ between 560 and 690 nm for the device embedding this optical design. Siegmund et al. used an MC effect to increase the normally negligible EQE in the CT absorption spectrum by more than 40 times.[133] In their system, electrons located

![Figure 7. a) Schematic representation of the device architecture of a resonant-cavity-enhanced organic BHJ photodetector. b) Architecture of a cavity-enhanced NIR detector; the combination of a dielectric DBR and OPD with a metal top mirror leads to the formation of Tamm plasmon-polariton states. c) Photodetector structure having a metasurface with period D. d) Diagram of the OPD structure and each layer’s material. e) Top left: Schematic representation and the cross-sectional SEM image of the device; top right: photograph of devices on a flexible substrate; bottom left: photograph reconstructed by combining images obtained from 2D scanning of artificial blue-, green-, and red-cone photodetectors over a photograph of a sunflower; bottom right: original photograph of the sunflower. f) Cross-sectional SEM image of an NIR detector, revealing its alternating LiF/NPB DBR structure. g) Schematic representation of a photodetector having an Ag resonance cavity. a) Reproduced with permission.[130] Copyright 2017, Wiley-VCH. b) Reproduced with permission.[131] Copyright 2017, American Chemical Society. c) Reproduced with permission.[132] Copyright 2018, American Chemical Society. d) Reproduced with permission.[205] Copyright 2020, Royal Society of Chemistry. e) Reproduced with permission.[133] Copyright 2019, Wiley-VCH. f) Reproduced with permission.[134] Copyright 2020, Wiley-VCH. g) Reproduced with permission.[135] Copyright 2020, Wiley-VCH.](https://www.advancedsciencenews.com/doi/10.1002/aes.2000035)
in an occupied state of the donor material were promoted to the lowest unoccupied molecular orbital of the acceptor, thereby efficiently dissociating the free carriers to their corresponding electrodes. Wang et al. achieved a continuously adjustable detection wavelength by varying the thickness of the transmission layer (hole transporting layer/active layer/ETL) between Ag mirrors. At the optimized active layer thickness, the photodetector displayed a noise-free monochromatic spectral response (FWHM: 25 nm; EQE: >50%). Gibert-Roca et al. used nanoimprint- ing to form the active layer of PC nanostructures; by changing the lattice parameters of the PC structure (Figure 7d), the spectral response could be adjusted to greater than 1000 nm. The response of the device at 900 nm was as high as 50 mA W⁻¹.

Artificial eye is an emerging application, especially in the fields of night vision, machine vision, and clinical applications. Advances in OPDs and PePDs have pushed the detectability of light signals beyond that of the human eye. An MDN MC electrode can act as a transmission color correction medium for determining the color-sensitivity of PD devices (narrowband PD). Tsai et al. reported a fully vacuum-processed perovskite photodetector integrating an MDN cavity to mimic the photore- sponse of human retina cells (Figure 7). Through careful manipulation of the width of the MC and the optical field distribution, detectors have been obtained displaying large LDRs (up to 154 dB), impressive detectivity (up to 10¹⁴ Jones), and spectral responses better than that of the human eye. Li et al. prepared a narrowbandwidth PePD by integrating an alternating Ag/ITO resonant cavity (Figure 7f). The transmittance spectrum of the cavity could be tuned by varying the number and the size of cavities. Single-cavity ITO/Ag/ITO with double-resonant ITO/Ag/ITO/Ag/ITO/Ag/ITO structures served as narrowband filters, while the three-resonator ITO/Ag/ITO/Ag/ITO/Ag/ITO/Ag/ITO/Ag/ITO structure provided a wider transmission bandwidth. By varying the thicknesses of the Ag and ITO layers, they fabricated color sensors centered at 465, 510, 600, and 635 nm, with LDRs (>139 dB) surpassing that of the human retina. Cao et al. reported an NIR-I CsPb₂Br₂SnO₃I₃ detector equipped with a butterfly-inspired LiF/NPB alternating-layered DBR (Figure 7g). By tailoring the thicknesses of the LiF and NPB layers, they realized photoresponse spectra located at 800, 850, and 900 nm. These wavelengths lie within the optical window for in vivo imaging and are particularly relevant for biomedical research. The devices displayed an FWHM of less than 50 nm, notable detectivity of up to 5.4 × 10¹⁴ Jones, and detection limits as low as 0.33 nW.

To realize high-efficient narrowband PDs, Burn coworkers introduced the concept of the charge-collection-narrowed (CCN) effect into OPDs. The CCN effect arises from a difference in absorption coefficients at short and long wavelengths. When the active layer is sufficiently thick, the carriers generated from photons with high extinction coefficient would mostly distribute near the illuminated surface and would not be collected by the circuit before their recombination. In contrast, photons with relatively low extinction coefficient could penetrate deep into the active layer; the photogenerated carriers would distribute uniformly in the device and would eventually be collected, contributing to the photocurrent and resulting in a narrowband photoresponse. Notably, the EQE would be quite low for this type of PD. This strategy provides filter-free structures for narrowband OPDs as well as PePDs. By tuning the halide composition of the perovskite and adding an organic molecule (e.g., rhodamine B), Lin et al. could independently control the extinction coefficients at long and short wavelengths, respectively. PePDs having tunable spectral responses and FWHMs of less than 100 nm were obtained.

Rao et al., Saidaminov et al., and Zhou et al. used thick perovskite layers to fabricate self-filtered PePDs, with Rao et al. using MAPbBr₃, Saidaminov et al. using MAPbX₃ (X = Cl, Br, I), and Zhou et al. using Cs₂SnCl₆–Brₓ, as their active layers. The PePD prepared by Rao et al. exhibited narrowband detection centered near 570 nm (FWHM = 35 nm) with a broad linear response from 10⁻⁴ to 10⁻² mW cm⁻², a high wavelength selectivity of 61.3 dB, and a 3-dB cutoff frequency (3dB) of 110 kHz under a low bias of ~1 V. The photoresponse of the perovskite photodetectors developed by Zhou et al. could be tuned from 400 to 600 nm, with an FWHM of 45 nm and a high detectivity of 2.71 × 10¹⁰ Jones. The photodetectors prepared by Saidaminov et al. had a narrow detection spectrum centered near 830 nm, with an FWHM of 33 nm, and a high EQE (>10⁶%).

7. Conclusions

Organic- and perovskite-based PVs and PDs have significant advantages over conventional inorganic counterparts—most notably flexibility, wet processability, low fabrication and temperatures suitable for large-scale manufacture, and tunable optoelectronic properties—that would be beneficial to a variety of novel applications. Although there has been much recent progress, as discussed earlier, there remain many challenges limiting the commercialization of these devices. Later, we discuss several key issues that must be addressed.

The main challenge is the development of emerging materials displaying efficient intrinsic optoelectronic properties, in terms of their molecular design and their integration with device optical engineering. For instance, state-of-the-art OPVs usually feature active layer materials having complex molecular structures. For perovskite materials, the deposition of high-performance perovskite layers remains a “black magic,” with the purity of the precursors, the temperature, the humidity, the surface energy of the substrate, and the composition of the perovskite all affecting the stability and performance of the devices, often varying from lab-to-lab (i.e., poor repeatability and reproducibility). Integration of additional optical structures leads to more issues, because they introduce new fabrication conditions (e.g., additional processing temperatures and atmospheres, underlayer wettability, and possible ionic and sputtering damage) to the original devices. There are two approaches to overcome these problems: 1) Manufacturing the optical structures before preparing the PVs and PDs. For those optical structures requiring high processing temperatures or harsh chemical environments, it would be preferable to fabricate them first and provide the same (or similar) underlayer for further deposition of the PVs and PDs. 2) Room-temperature solvent-free fabrication. Some of the optical structures may be required to be made after the deposition of the PV and PD layers. If that is the case, the optimized active layers of the PVs and PDs will have already been made. The additional optical structures will have to be added without
altering the already-optimized devices. Hence, keeping the substrates at room temperature and not subjecting them to further solvent-soaking would be crucial. Some deposition techniques (e.g., solvent-free vacuum sublimation or film laminating) would be more suitable than solution processing, which might, however, be most suitable in some instances. Some special care (e.g., using a protection layer or an orthogonal solvent system) might be necessary, with the knowledge that such methods might introduce additional optical effects to the final devices.

From the point of view of ST-PVs, materials are still being sought with preferred NIR and UV light-harvesting ability. The color and transparency of the devices can be altered by applying some of the optical designs (MC, DBR, and PC) mentioned herein, with selective transmission or improved specific reflection to reharvest originally unabsorbed light. For complex device architectures, powerful simulation tools (e.g., artificial intelligence or deep mind learning) might be helpful to provide device architectures, powerful simulation tools (e.g., arti-

The color and transparency of the devices can be altered by applying some of the optical designs (MC, DBR, and PC) mentioned herein, with selective transmission or improved specific reflection to reharvest originally unabsorbed light. For complex device architectures, powerful simulation tools (e.g., artificial intelligence or deep mind learning) might be helpful to provide guidelines to facilitate the design of ST-PVs. Today’s preparation technologies still rely heavily on vacuum (sputtering and evaporating) deposition electrodes or mirrors to realize the designed optical structures. The reliability of solution-processed nanowires, nanotubes, and conducting polymers remains questionable. The development of highly transparent and conductive materials with high processability for the preparation of transparent electrodes remains urgent. There are also great opportunities for embedding specially designed optical structures in next-generation PDs; such research is seldom performed, as is evident in this Review. Furthermore, studies into the optical engineering of low-noise IR detectors (at room temperature), ionizing radiation detectors, photomultiplication detectors, and photodetector systems and arrays are also rare. Overall, the development of PDs based on organic and perovskites remains an exciting field. With continued advances in optical engineering, next-generation photodetectors will surely appear.

Finally, the main obstacle inhibiting the commercialization of these new devices is their long-term stability. Research groups worldwide are making great efforts to investigate the degradation mechanism of these materials and the possible solutions. Most of the devices described herein were characterized in glove boxes at room temperature. Device performance and stability under realistic and/or harsh testing conditions are rarely investigated or reported. The encapsulation of these optoelectronic devices should be considered to prolong their lifetimes. In particular, BIPV applications require a lifetime of at least 5–10 years. The effects of encapsulation on the optical properties of the devices will also need to be investigated, with a prerequisite of substantially improved stability. All these challenges remind us the necessity for innovation and inspiration, to polish the manufacturing techniques of the future.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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