Photonic crystals for perovskite-based optoelectronic applications

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Abstract
The emerging perovskite materials present great opportunities for cost-saving and efficient optoelectronic devices. However, there are still some roadblocks in the path toward commercialization of perovskite-based devices, such as low light utilization, poor crystalline quality and dissatisfactory environmentally long-time stability. To solve these problems, photonic crystal (PC) as a promising structure for control light has been exploited. To date, a series of perovskite-based devices improved by PC has been reported. These PCs do not only improve the performance of devices but also give flexural endurance, vivid color and some degree of transparency to the electronic devices. Herein, a general overview is provided on the recent advances for the application of PCs in perovskite-based optoelectronic devices (solar cells, photodetectors, and light-emitting diodes). In addition, some personal perspectives on potential future challenges and improvements in this field are presented.

KEYWORDS
light-emitting diodes, perovskite optoelectronic materials, photodetectors, photonic crystals, solar cells

1 INTRODUCTION

Due to the excellent optoelectronic properties such as ambipolar charge transport, long carrier diffusion length (> 1 μm in perovskite polycrystalline films), tunable optical bandgaps, considerably high carrier mobility (~10 cm² V⁻¹ s⁻¹), and excellent photoluminescence quantum yield, perovskite (ABX₃: A = CH₃NH₃ (MA), HC(NH₂)₂ (FA), Cs, Rb; B = Pb, Sn; X = I, Br, Cl) crystal has triggered enormous attention as emerging photoactive materials in optoelectronic applications.¹⁻⁹ Meanwhile, the low-cost raw materials, facile solution processability, and flexibility of perovskite materials enable these very promising for commercialization in the flexible devices.¹⁰⁻¹²

However, the issues of light utilization, interfacial properties, and long-time stability of perovskite-based devices are the bottlenecks for future practical applications. Moreover, the crystalline quality is the Achilles’ heel for manufacturing scalable perovskite films. To overcome those problems, a myriad of methods including architecture optimization,¹³ chemical doping,¹⁴⁻¹⁹ functional material design,²⁰,²¹ and interface engineering²²⁻²⁷ have
been exploited. In particular, the beneficial effects of optical designs in the perovskite-based optoelectronic devices have been proved. A suitable optical design can simultaneously enhance the light trapping (LT), prolong the optical path length, improve the perovskite film quality and release the residual stress. \[28–34\]

Photonic crystal (PC) is one of the most common bio-inspired ordered periodic structures for manipulating light.\[35–38\] According to the structure of PC, it can be divided into one-dimensional (1D) PC, two-dimensional (2D) PC, and three-dimensional (3D) PC, as shown in Figure 1. The variety of color by PC is attributed to its periodically arrayed photonic structures which have the capability of manipulating the diffraction and/or reflection of light.\[39,40\] The light propagation is forbidden in the featured frequency range by periodic change of refractive index (PC constituents) that engenders a forbidden gap in the photonic band structure. Thus, the PC exhibits many unique optical properties such as photon localization, fluorescence enhancement effect, and “low-photon” effect.\[41–45\] Based on the above properties, PC receives widespread attention and shows great potential applications in numerous fields, such as lasers,\[46–48\] anti-counterfeiting technologies,\[49\] smart detectors,\[50–56\] catalysis,\[57–59\] blue phase liquid crystal display,\[60,61\] light-emitting diodes (LEDs),\[62–64\] and solar cells.\[65–72\] Particularly, the applications of PC in the perovskite-based optoelectronic files have been rapidly developing in recent years.

In this review, we collect some representative examples of perovskite-based optoelectronic devices (solar cells, photodetector, and LEDs) with PC. The relationships between the PC structures and the improved efficiency as well as enhanced stability of perovskite-based optoelectronic devices are also discussed. Finally, the summary and outlook of the future development of PC in perovskite-based optoelectronic devices are proposed.

2 | APPLICATIONS OF PC IN PEROVSKITE-BASED OPTOELECTRONIC DEVICES

2.1 | PC in solar cells

Perovskite solar cells (PSCs) as the promising next-generation photovoltaic devices have been rapidly developing in the past decade years and the power conversion efficiency (PCE) of PSCs is soaring from 3.8% in 2009 to beyond 25.5% \[10,73–84\] Owing to the deficient light utilization of currently PSCs, the performance of PSCs is hard to be improved despite that the theoretically Shockley-Queisser limit is 33%.\[85\] Light absorption is the crucial property of PSCs that generate photocurrents. It is certified
that the light utilization of the light-related devices is largely dependent on the micro-/nanostructure of each functional layer on the devices.[86–88] Usually, the architecture of PSCs is a planar structure. However, the light propagation path of the planar structure is single, which greatly limits the light absorption performance of devices. PC with novel micro-/nanostructures is a potent approach to increase light utilization, thereby accomplishing a high-performance device.[28,89]

Many researchers focus on designing novel PC structures into each functional layer of PSCs to improve the performance of devices. Zhou et al. reported a carbon quantum dot (CQD) sensitized CsPbBr3 inverse opal (CQD/CsPbBr3 IO) films as the light active layer for PSCs. Figure 2A(a, b) showed the configuration of the CQD/CsPbBr3 IO PSCs and the scanning electron microscopy (SEM) images.[90] The photonic band gaps (PBG) of CsPbBr3 IO can be confirmed by the extra absorption peak of 670 nm that appeared in the IO sample. The slow photon effect was introduced by the CsPbBr3 IO films to increase the path length of light, which can greatly enhance the light utilization of devices. Moreover, comparing with planar CsPbBr3, the light absorption intensity of IO-structured samples was significantly enhanced in the whole spectrum. This is mainly attributed to the optimized light harvesting ability, the enhanced light interaction, and multiple incident light scattering among the highly periodically 3D macroporous IO structures. Finally, the PCE of PSCs based on the CQD/CsPbBr3 IO films improved to 8.29%, which is two times higher than that of planar devices without IO films, as shown in Figure 2A(d).

Apart from the perovskite film, some novel PC structures incorporated in the transporting layer of the PSCs have also been explored to maximize light absorption and achieving a high-performance device. Choi et al. designed a 2D TiO2 PC nanodisk array as the electron transport layer for the PSCs by nanosphere lithography (Figure 2B(a)).[32] Compared with the pristine sample, the 2D TiO2 nanodisk array showed a lower transmittance in the wavelength range of 600–750 nm, which can be assigned to the photon confinement effects of the nanodisk array and the enhanced light-harvesting (Figure 2B(B)). Besides, the author analyzes the optical responses of the nanodisk array through Finite-difference time-domain (FDTD) simulation, the resulting electromagnetic energy density distributions have been shown in Figure 2B(c and d). The nanodisk sample versus pristine film is turning higher light restriction and forward scattering. As expected, the PSCs with 2D TiO2 nanodisk array exhibited a higher Jsc and PCE. This was mainly ascribed to the stronger light scattering from the nanodisk array in comparison with the device with flat TiO2. Notably, Haque et al. presented two types of TiO2 LT structures with anti-reflection and light scattering properties, which were used as the electron transporting layer (ETL) in the PSC. The device constructions were shown in Figure 2C(a and b).[31] Owing to the broadband light absorption gain from the LT features, the device based on TiO2 LT structure has a pronounced increase in the photocurrent than that of without LT structure and allows reducing the perovskite layer thickness without influence to performance (Figure 2C(c)). The improvements of photocurrent are close to the predicted value from theoretical lambertian limits. Additionally, the ultraviolet blocking property of TiO2 with LT structures can lead to an enhanced photostability of the PSCs (Figure 2C(d)). The nanocellular transport layer scaffold featuring an IO structure can also be employed to construct a mechanics buffer layer and optics resonant cavity. For example, Hu et al. fabricated a polystyrene-doped nanocellular PEDOT:PSS to boost the performance and mechanical robustness of the device.

The configuration of the PSCs and the SEM image of the nanocellular PEDOT:PSS layer were shown in Figure 2D(a and b).[91] The refinement of light-harvesting capability by the optical resonant cavity of the nanocellular charge transporting layer was greatly improved. The devices based on the nanocellular scaffold exhibited a higher PCE of 36% than that of the control devices. Particularly, as illustrated in Figure 2D(c and d), a high flexural endurance PSCs was achieved, owing to the mechanical stresses releasing function of the nanocellular scaffold. Besides improved efficiency and flexural endurance, the PC applied in PSCs can also achieve other beneficial effects, such as vivid color. The tunability of color is a particularly appealing add-on property for perovskite solar cells because aesthetical versatility can expand its application to building-integrated photovoltaics (BIPVs) and wearable devices. Despite the efficiency of PSCs increased rapidly, the color gamut available of PSCs was limited and did not cover the green-to-blue region of the visible spectrum. To broaden the color gamut of PSCs, Zhang et al. prepared a porous PC scaffold via alternating liquid processed layers of dense titanium dioxide (TiO2) and porous silicon dioxide (SiO2), which was integrated as a substitute scaffold in PSCs.[92] As shown in Figure 3A, the author optimized the porosity of each layer in the PC for maximized the reflectance of the device to obtain the colorful PSCs. The structural color by photonic structure is more stable than that of doping colorful absorber materials. In addition, the colorful PSCs with PC scaffold for BIPV exhibit a decrease of thermal gain in the PSCs and building owing to the light reflection property of PC, which is beneficial for increasing the efficiency of PSCs in the ambient conditions and reducing the cooling demand in buildings. Transparency is an important factor in the application of PSCs. To form the colored semi-transparent
FIGURE 2 PCs integrated into perovskite solar cells (PSCs) to improve performance. A, (a) The configuration of the CQD/CsPbBr$_3$ inverse opal (IO) PSCs.\textsuperscript{[90]} (b) The SEM images of CQD/CsPbBr$_3$ IO.\textsuperscript{[90]} (c) Schematic diagram of the reflection experiment.\textsuperscript{[90]} (d) Current density-voltage ($J-V$) curves of PSCs based on CsPbBr$_3$ planar, CQD/CsPbBr$_3$ planar, CsPbBr$_3$ IO, and CQD/CsPbBr$_3$ IO.\textsuperscript{[90]} Copyright 2017, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. B, (a) The configuration of the PSCs with 2D PC nanodisk (ND) array ETL.\textsuperscript{[32]} (b) The enlarged UV–Vis transmission spectra of perovskite-coated, pristine, and 2D ND arrays.\textsuperscript{[32]} (c, d) Electromagnetic energy density distributions of electromagnetic waves diffracted by the TiO$_2$ layer with 2D ND array morphologies.\textsuperscript{[32]} Reproduced with permission.\textsuperscript{[32]} Copyright 2019, Elsevier. C, (a-b) Sketch of both types of LT structures, composed of TiO$_2$ features integrated on the ETL of the PSCs.\textsuperscript{[31]} (c) Absorption spectra of the PSCs with optimized LT structures.\textsuperscript{[31]} (d) The analysis of photocurrent enhancements with the optimized photonic structures.\textsuperscript{[31]} Reproduced with permission.\textsuperscript{[31]} Copyright 2019, Elsevier. D, (a) The configuration of the PSCs with the nanocellular (NC) PEDOT:PSS layer.\textsuperscript{[91]} (b) SEM image of the NC-PEDOT:PSS layer.\textsuperscript{[91]} (c, d) Scheme of stresses release by the NC-PEDOT:PSS structure.\textsuperscript{[91]} Reproduced with permission.\textsuperscript{[91]} Copyright 2017, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.
PSCs, Lee et al. used an e-beam evaporator to prepare a multilayer dielectric mirror that consists of SiO$_2$ and TiO$_2$ as highly reflective 1D PC upon the anode of the device. Figure 3B was shown the schematic structure of the colored semi-transparent PSCs.\textsuperscript{93} The number of pairs of alternating layers in the dielectric mirror and the angle of incidence has been optimized to broaden the color gamut of the device. The optical simulation results show that the optical band gap effect of the 1D PC can improve the light absorption efficiency of the semi-transparent device and maintain certain transparency. The resulting PSCs have enhanced light harvesting due to the strongly reflected of light beyond 550 nm wavelength back to the perovskite layer. However, the baseline ripple-shape patterns were induced by the secondary interference effect in 1D PC can lead to a decrease of color purity and maybe creates an unwanted reflection which reduced the PCE of PSCs. The PSCs with the above porous PC scaffold showed a PCE of 7.9\%, which was reduced 23\% than that of the mesoporous SiO$_2$ as a scaffold. Recently, Yoo et al. proposed a nonperiodic 1D PC which was comprised of multiple alternating layers of low-index SiO$_2$ and high-index TiO$_2$, as shown in Figure 3C(a).\textsuperscript{94} The baseline ripple-shape pattern was nearly eliminated by the nonperiodic 1D PC (Figure 3C(b)). Under AM 1.5G illumination, the colorful device with the nonperiodic 1D PC exhibited an optimized PCE of 18.9\% while the control PSC had a PCE of 20.1\%, reaching a PCE loss only of 7\% (Figure 3C(c)).

## PC in photodetector

The photodetector refers to a semiconductor device that can observe extremely weak optical signals and converts them to electrical signals. They have a wide range of applications in optical communication, medical analysis, environment monitoring, security systems, and so forth. The structural thickness is a crucial role in photon absorption of the photodetector, which determines the detecting performance and manufacturing cost. Perovskite materials have attracted great interest in photodetector devices owing to their high external quantum efficiency (EQE),
low trap-state density, and long diffusion length. The reflection and crystallinity of the perovskite layer are vital to the light-harvesting and efficiency of photodetectors.

It is well known that the light-harvesting of optoelectronic devices can be improved by PC structure. Inspired by the hierarchical architecture of the butterfly, Zhan et al. designed a perovskite-based photodetector with 1D nanograting bonded porous 2D PC (G-PC) perovskite film via a facile and reproducible method. The 2D crystalline colloidal template by air-liquid self-assembly was employed to obtain porous 2D PC perovskite film, and the grating structure was constructed by nanoimprinting with a commercial DVD master, as shown in Figure 4A. The author used 3D FDTD method to calculate the field intensity distribution in perovskite films with various optical structures. The results showed that the G-PC structure facilitated the light propagation into a deep perovskite active layer, while the intensity decreases sharply for a planar structure. The photodetector with G-PC exhibited a greatly improved light-trapping of the active layer and reduced light reflection to enhanced light-harvesting of the device. Compared with the planar perovskite device, the white light responsivity and detectivity of the photodetector with G-PC were increased, which was up to six to seven times. Yang et al. synthesized a CsPbBr$_3$ single crystal, the photodetector based on the single-crystal exhibited high responsivity. However, the size and thickness of perovskite single-crystal can be quite different due to the solution method. Besides, the low solubility of CsBr in present solvents limited the formation of high-quality CsPbBr$_3$ film by solution process. To solve the problem, Zeng et al. fabricated an ultradense CsPbBr$_3$ IO film with preferred orientation and enlarged crystal grains by a space-confined growth strategy. The corresponding top-view SEM image was shown in Figure 4B(b). The photodetector based on the CsPbBr$_3$ IO films exhibited high sensitivity and short response time within 5 seconds. The sensitivity of 216 A W$^{-1}$ was 40 times over that of the photodetector with flat films. All-inorganic perovskite nanocrystals (NCs) and quantum dots (QDs) exhibit high emission efficiency and broadly tunable bandgap energies, thus also considered for photodetector. For instance, CsPbCl$_3$ NCs with a bandgap of 2.82 eV were one of the ideal choices for photodetection materials. However, the low quantum yield of CsPbCl$_3$ NCs caused by the high density of deep defect states (the Cl vacancy) limited the detection sensitivity of the device. Recently, Li et al. firstly reported an approach that combing the Ag plasmon and PC to enhance the luminescence yield of NCs. The schematic of the CsPbCl$_3$ on Ag/PCs photodetector was shown in Figure 4C(a). The resulting luminescent intensity of CsPbCl$_3$ NCs boosted more than 150-fold due to the effects of surface plasmon and PC (Figure 4C(b)). The photodetector with CsPbCl$_3$/Ag/PC afforded an exceedingly low dark current of 10$^{-11}$ A and narrow response line width of 30 nm. It also showed an ultrashort response time of 28 ms/31 ms and a high detectivity of 9 × 10$^{13}$ Jones, which was significantly enhanced 40–50% and 682%, respectively compared with the traditional CsPbCl$_3$ photodetector (Figure 4C(c)).

Furthermore, stability as an important parameter to assess the photodetector has also attracted a lot of attention very recently. Chen et al. innovatively demonstrated high stability and sensitivity photoelectrochemical (PEC) detector composite by 3D TiO$_2$ PC, CsPbBr$_{1.5}$I$_{1.5}$ QDs, and Nafion protective membrane. The schematic illustration of the principles of PEC detection under visible light excitation was shown in Figure 5A(a). The existence of porous 3D TiO$_2$ PC has significantly improved the stability of the PEC detector because of its high specific surface area and mass active sites. Meanwhile, the photocurrent response of the detector based on TiO$_2$ PC/CsPbBr$_{1.5}$I$_{1.5}$ QDs was enhanced which attributed to the photonic stop band effect of TiO$_2$ PC (Figure 5A(b)). Under the light illumination of 600 nm, the photocurrent response of the device with PC/QDs only showed a 7% decrease after 3 hours. In comparison, the detector based on QDs exhibited a 30% decrease in the same condition. The nanocellular structure was also utilized in the photodetector to enhance the stability of the device. In Figure 5B, Tian et al. engineered a nested nanocellular flexible and self-driven perovskite photodetector via a facile template-assisted spin-coating method. The SEM image of the nanocellular perovskite film was shown in Figure 5B(b). The nanocellular perovskite film exhibited a lower trap density of 3.4 × 10$^{15}$ cm$^{-3}$ compared with the planar structure (5.8 × 10$^{15}$ cm$^{-3}$). The photodetector with the high crystallization nanocellular perovskite film was achieved a high responsivity of 473 mA W$^{-1}$ and detectivity of 1.35 × 10$^{13}$ Jones. Importantly, the nanocellular device exhibited excellent bending repeatability and long-term stability in an atmospheric environment. The finite element simulation was used to probe the mechanical stress distribution of the nanocellular and planar perovskite film. Simulation results demonstrated the stress distribution of nanocellular film with higher homogeneity than the planar film and less prone to collapse (Figure 5B(c and d)).

### 2.3 | PC in LEDs

While perovskite materials have shown an excellent performance in the solar cell, they can also be attractive materials for LEDs. The parameters for evaluating the performance of the LED mainly included photoluminescence quantum yield (PLQY), EQE, luminance or brightness,
FIGURE 4  PCs integrated into perovskite photodetector with increased sensitivity and responsivity. A, Illustration of the fabrication procedure. Reproduced with permission. [95] Copyright 2019, American Chemical Society. B, (a) Schematic illustration of conventional and space-confined CsPbBr₃ film fabrication strategies. [34] (b) Top-view SEM image of a CsPbBr₃ IO film with a large area. [34] (c) Responsivity spectra under a bias of 1 V. Reproduced with permission. [34] Copyright 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. C, (a) Schematic of the CsPbCl₃ on Ag/PCs photodetector. [97] (b) Comparison between emission spectra of CsPbCl₃ film and CsPbCl₃/Ag/PC. [97] (c) On-off switching properties measured under 365 nm light illumination (10 mW cm⁻²) at a bias of 3 V. Reproduced with permission. [97] Copyright 2018, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim
and current efficiency (CE). Normally, the perovskite-based LEDs have a similar configuration with the PSCs, which are comprised of a perovskite light-emitter layer and two types of transporting layer. The operation of LEDs is involved in the following processes: the charge carriers transported to the perovskite emitter layer across the electrode for efficient radiative recombination. Thus, the methods for improved perovskite-based LEDs efficiency should be focused on the three key aspects: balanced injection of electrons and holes, minimal nonradiative recombination loss, and high extraction of emitted photons. The EQE of perovskite-based LEDs has been improved from less than 1% reached to more than 20% in only 4 years.\textsuperscript{100} However, EQE in conventional perovskite-based LEDs remains limited by the low light extraction and outcoupling efficiency. Recently, Zhang et al. obtained an optimized perovskite-based LEDs device with high performance through the anodic alumina membranes (AAMs) as a 3D nanophotonic substrate.\textsuperscript{101} (Figure 6A(a)) The AAMs nanophotonic substrate was composed of the nanodome (ND) arrays light coupler and the nanowire (NW) arrays optical antennas. The SEM image of AAMs has been shown in Figure 6A(b). The light extraction of CH$_3$NH$_3$PbBr$_3$ (Br-Pero) with AAMs was enhanced substantially and can be achieving a highly EQE of 17.5%, which was about two times increase than the planar control device (Figure 6A(c)). Furthermore, Shen et al. presented a facile route to fabricate bioinspired moth-eye nanostructures (MEN) to enhance the outcoupling efficiency of waveguided light in conventional LEDs, as shown in Figure 6B(a).\textsuperscript{102} Compared with the flat film, the substrates with MEN pattern showed a remarkable increase in optical transmission and haze at the broad spectral range of 350–800 nm, which represented the stronger light scattering of pattern film and can lead to a reinforce light outcoupling of corresponds LED (Figure 6B(b)). As a result, the MEN-based LED yielded a superior EQE of 20.3% and CE of 61.9 cd A$^{-1}$, which were 1.5 times for the flat one.
**FIGURE 6**  PCs integrated into perovskite LEDs with enhanced EQE. A, (a) Schematic the LEDs on the nanophotonic substrate. The materials from top to bottom are: Ca/Ag electrode, poly(9,9'-dioctyl-fluorene), CH$_3$NH$_3$PbBr$_3$ (Br-Pero), PEDOT:PSS, ITO, and AAM. AAM channels are filled with TiO$_2$.\[101\] (b) SEM image of the barrier side of the free-standing AAM film with nano domes structures.\[101\] (c) EQE enhancement factor of AAM devices compared with the thin film control device. Reproduced with permission.\[101\] Copyright 2019, Springer Nature. B, (a) The configuration of the CsPbBr$_3$ LED with the imprinted nanostructures.\[102\] (b) Total transmittance and haze of flat and patterned substrates. Reproduced with permission.\[102\] Copyright 2019, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

**3 SUMMARY AND OUTLOOK**

In summary, this perspective focuses on the applications of PC in perovskite-based photoelectric devices including solar cells, photodetector, and LEDs. The PC showed the excellent ability to manipulate the reflectance, scattering, transmittance, and absorption of film. Thus, the light utilization of perovskite-based photoelectric devices can be significantly improved after incorporated with PC. Besides, the mechanical stability and environmental stability of devices were also enhanced due to the uniform stress distribution and hydrophobicity of the PC layer, which was expanded the commercial application prospect of perovskite-based photoelectric devices. Although the performance of the perovskite-based photoelectric devices incorporated with PC exhibited a significant enhancement, the path of commercial applications still has a long distance to reach. Several challenges are listed below:

1. The mainly building block materials of PCs applied in perovskite-based photoelectric devices remain limited to SiO$_2$ and TiO$_2$. To further improve the performance of devices, such as the transmittance of colorful PSCs, the sensitivity and responsivity of perovskite photodetector, the EQE of perovskite LEDs, it is necessary to exploit more efficient PCs with new building block materials and stable structures.

2. Regardless of the effectiveness of PC in perovskite-based photoelectric devices, the large-area PC film with an acceptable cost is still a challenge. Air-liquid self-assembly is the common method to prepare PC, but the uniformity of large-area PC film still needs to be improved. Photolithography and e-beam evaporator can be used to fabricate large-area PC with high uniformity, however, the cost is expensive. The trade-off between the cost and the quality in large-area PC limits the practicability of the perovskite-based photoelectric
devices with PC. From the view of industrial manufacturing process, the development of low-cost fabrication techniques to realize high-quality PC film is a pursuit that needs continuous efforts.

3. It is a future direction to combine different PC schemes for high-performance perovskite-based photoelectric devices. Particularly, the efficiency of solar cell devices with PC still has a large room for improvement by optimizing the light utilization in solar cell devices. Furthermore, PC can also be utilized to improve crystallization growth and charge transfer properties of the functional layer in PSCs. The combination of suitable PC structures should be considered.

Furthermore, the investigation about the PC in perovskite-based devices is still mainly focused on the improvement of optical performance for high effectiveness. However, the electrical property is also important to achieve high-effective photoelectric devices. Future exploration should focus on balance the optical and electrical properties in devices with PC. Progress about the PC in optoelectronic devices to achieve the transition of perovskite-based optoelectronic devices from bench to market needs to be investigated at a greater depth.

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CONFLICT OF INTEREST
The authors declare no conflict of interest.

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