All-inorganic halide-perovskite polymer-fiber-photodetector for high-speed optical wireless communication

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Abstract: The use of optical carrier frequencies will enable seamless data connection for future terrestrial and underwater internet uses and will resolve the technological gap faced by other communication modalities. However, several issues must be solved to propel this technological shift, which include the limitations in designing optical receivers with large detection areas, omnidirectionality, and high modulation bandwidth, mimicking antennas operating in the radio-frequency spectrum. To address this technological gap, herein, we demonstrate halide-perovskite-polymer–based scintillating fibers as a near-omnidirectional detection platform for several tens-to-hundreds of Mbit/s optical communication in both free space and underwater links. The incorporation of all-inorganic CsPbBr3 nanocrystals by engineering the nanocrystal concentration in an ultraviolet-curable polymer matrix ensures a high photoluminescence quantum yield, Mega-Hertz modulation bandwidth and Mbit/s data rate suitable to be used as a high-speed fibers-based receiver. The resultant perovskite polymer-based scintillating fibers offer flexibility in terms of shape and near-omnidirectional detection features. Such fiber properties also introduce a scalable detection area which can resolve the resistance-capacitance and angle-of-acceptance limits in planar-based detectors, which conventionally impose a trade-off between the modulation bandwidth, detection area, and angle of view. A high bit rate of 23 Mbit/s and 152.5 Mbit/s was achieved using an intensity-modulated laser for non-return-to-zero on-off-keying (NRZ-OOK) modulation scheme in free-space and quadrature amplitude modulation orthogonal frequency-division multiplexing (QAM-OFDM) modulation scheme in an underwater environment, respectively. Our near-omnidirectional optical-based antenna based on perovskite-polymer-based scintillating fibers sheds light on the immense possibilities of incorporating functional nanomaterials for empowering light-based terrestrial- and underwater-internet systems.

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1. Introduction

Optical-based wireless communication (OWC), which utilizes the unlicensed spectrum of 400 THz to 1500 THz, has been coined as a paradigm-shifting technology for fifth-generation (5G) communication networks and beyond because of its much larger bandwidth capacity, high resilience to electromagnetic interference, and low latency rate [1,2]. This disruptive technology will relieve the ever-increasing network congestion in the current radio frequency (RF) mobile network and to support various communication modalities in the absence of copper or fiber-optic networks, for example, vehicle-to-vehicle communication [3], underwater wireless
optical communication (UWOC) [4,5] and last-mile internet connectivity for remote communities [6]. However, while propelling the paradigm shifting technology, the photodiodes or avalanche photodiodes (APDs) used on the receiver end are typically limited in the active area to minimize the resistance–capacitance (RC) limit to enable a higher modulation bandwidth and warrant high-speed optical communication link in the range of at least tens of Mbit/s. For instance, the diameters of InGaN-based photodiodes are often restricted to less than 100 µm for a –3-dB frequency bandwidth \((f_{-3dB})\) of approximately 40 MHz to 347 MHz [7–9]. With such a large modulation bandwidth, although it is possible to achieve Mbit/s to Gbit/s optical communication link, such limitations impose strict pointing, acquisition, and tracking (PAT) requirements on the optical communication link, thus leading to the requirement of integration with costly and complex mechanical tracking systems [10]. Meanwhile, solar cells based on silicon [11,12], organic [13] and perovskite [14] material systems have also been proposed for large-area detection in OWC [15]. However, a low modulation bandwidth in the range of tens to hundreds of kHz is typically reported because of the RC limitation as a result of the large active area and thus restricts its application in achieving high-speed Mbit/s optical communication link.

In principle, optical elements such as lenses and compound parabolic concentrators (CPCs) can be integrated externally to improve the geometrical and optical throughput at the receiver end. However, these systems are limited by the fundamental principle of conservation of étendue, that is, \(A_{\text{in}} \Omega_{\text{in}} = A_{\text{out}} \Omega_{\text{out}}\), where \(A_{\text{in}}, A_{\text{out}}\) and \(\Omega_{\text{in}}, \Omega_{\text{out}}\) represent the active area and field-of-view solid angles of the optical element at the input and output, respectively [16–18]. This limitation imposes a trade-off between increasing the optical gain, which is defined by the ratio of \(A_{\text{in}}\) and \(A_{\text{out}}\), and expanding the field of view. Importantly, these optical elements are not suitable for omnidirectional detection owing to their rigid structures.

Recently, luminescent-concentrator–based optical receivers have been reported for OWC links, for example, luminescent-polymer–based fibers [19,20] and fluorescent slab antennas [16,18,21], which are not constrained by the étendue limit. The operation principle is similar to a luminescent solar concentrator (LSC) originally conceived for solar cell applications in the 70s [22], in which luminescent particles doped within a material with a high refractive index absorb incoming photons with a shorter wavelength \(\lambda_1\) and re-emit at a longer wavelength \(\lambda_2\), that is, \(\lambda_1 < \lambda_2\). In contrast to planar-based optical receivers, luminescent particles can absorb modulated optical signals independent of the incidence angle. The down-converted light is then guided via total internal reflection to a compact photodiode for optical-to-electrical (OE) conversion. These structures offer scalable, near-infinite optical gain by increasing the input active area \(A_{\text{in}}\) [21], while allowing a near-omnidirectional field of view suitable for signal detection in an OWC link using non-planar geometries [19]. The frequency bandwidth of the aforementioned luminescent concentrators is governed by the radiative recombination lifetime \(\tau\) of the luminescent particles, i.e., \(f_{-3dB} \leq 1/2\pi\tau\) [23,24]. Recent studies on luminescent-concentrator–based receivers for OWC links have thus far focused on standard organic-based luminescent materials, for example, coumarin 6 (Cm6) [16,18], 4-(dicyanomethylene)-2-methyl-6-(4-dimethylaminostyryl)-4H-pyran (DCM) [18] and SuperYellow [21]. Limited studies have focused on emerging luminescent materials based on halide perovskite material systems. Halide perovskites (ABX₃, where \(X = \text{Cl, Br, or I}\)) have emerged as a new class of materials for several optoelectronic devices and applications, for example, solar cells [25], light-emitting diodes [26], photodetectors [27,28], as well as phosphor conversion layers for solid-state lighting and OWC systems [29] because of their ease of solution processability and wavelength tunability covering the range from visible light to the near-infrared spectrum. In particular, all-inorganic CsPbBr₃ nanocrystals (NCs) with high photoluminescence quantum yields (PLQYs) of near unity and fast radiative recombination lifetimes in the range of less than 10 ns are suitable as fast phosphor-conversion materials for visible laser-based transmitters [29] and UVC-based receivers [30]. Although several prior studies have reported planar perovskite-based detectors targeting OWC applications [31,32],
these devices use less favorable planar-coated active layers. Therefore, they have limited étendue limits and are susceptible to RC limitations when scaling with the detection area.

Herein, we demonstrate a perovskite NC-polymer-based luminescent fiber for a large detection area and near-omnidirectional detection in an OWC link, which can overcome both the étendue and conventional RC limits. Our facile approach of adopting perovskite NCs into the polymer-based fiber ensures scalability and flexibility to suit various targeted needs in the OWC link, as compared to the planar structure. The perovskite-polymer luminescent fibers formed based on injection-molding and curing methods were optimized in terms of the NC concentration to ensure sufficient optical gain by reducing reabsorption losses. Embodiment of the perovskite NCs within the ultraviolet (UV)-curable polymer and polymer-based fiber frameworks also provides additional passivation in ambient environments for future internet-of-things (IoT) and internet-of-underwater-things (IoUT) systems. Small-signal modulation measurement based on a 405-nm laser diode transmitter confirmed the feasibility of high-frequency modulation with a proof-of-concept perovskite NC-polymer fiber, which is scalable for large-area detection if arranged into a flexible fiber array form. Notably, our demonstration sheds light and paves the way towards the transmission of hundreds of Mbit/s through OWC links based on the scalability and omnidirectionality of the data detection scheme, which promises a more robust interlinked ad hoc network.

2. Materials and methods

2.1. Synthesis and characterization of CsPbBr$_3$ NCs

High-quality CsPbBr$_3$ NC solutions were obtained commercially from Quantum Solutions. The NC solution was synthesized using a modified hot injection method. The NCs contained both oleic acid (OA) and oleylamine (OLA) as capping ligands on the surface [33]. The as-synthesized NCs were then mixed with the monomer, that is, isobornyl acrylate (IBOA), and a photoinitiator (PI) with an NC concentration of 20 mg/mL. For transmission electron microscopy (TEM) and X-ray diffraction (XRD) characterization, the NC samples prepared using the same method were dispersed in toluene for ease of solvent evaporation. TEM images were acquired using an FEI Titan ST system operating at 300 kV. Diluted CsPbBr$_3$ NCs with a concentration of approximately 1 mg/mL were drop-casted onto a carbon-coated 300 mesh copper TEM grid and subsequently dried in a vacuum chamber to completely remove the solvent. XRD characterizations were performed using a Bruker D2 Phaser based on Cu $K\alpha$ radiation ($\lambda = 1.5405$ Å). The photoluminescence (PL) spectra of the samples were measured using a 405-nm diode-pumped solid-state (DPSS) laser as the excitation source and collected using a CCD-based spectrometer (Ocean Optics, QE65000 Pro). The absorption spectra were collected using a UV-VIS-NIR spectrophotometer (Shimadzu, UV-3600). Time-resolved photoluminescence (TRPL) measurements were performed using a time-correlated single-photon counting (TCSCP) spectrophotometer (Horiba, FluoroMax 4). All measurements were performed in an atmospheric environment at room temperature.

2.2. Fabrication of CsPbBr$_3$ NCs-polymer fibers

The cladding layer of the polymer-based fiber made of polydimethylsiloxane (PDMS) was fabricated by mixing the elastomer base and curing agent at a ratio of 10:1. It was then cured at an elevated temperature of 80°C for ~1 h using a plastic mold. A fine needle was fixed at the center of the plastic mold to form a hollow core structure. After the PDMS was cured, the as-prepared CsPbBr$_3$-IBOA composite was injected into the core structure and subsequently cured under a UV light source ($\lambda_{\text{peak}} = 365$ nm) for 20 min. The polymer-based fiber was then be cleaved using a sharp polymer cutter.
2.3. Frequency bandwidth measurement

A 405-nm laser diode (Nichia, NDV4316) was mounted onto a customized thermoelectric cooler (TEC) (SaNoor Technologies, SN-LDM-T-405) and connected to a vector network analyzer (Agilent, E5061B) through a bias-tee (Mini-Circuits, ZFBT-4R2GW-FT+). The optical beam was collimated and collected using a perovskite-polymer fiber-based detector at the receiver end. The down-converted light escaping the fiber end was guided through a 450-mm–long silica-based fiber with a core diameter of 600 µm (Thorlabs, FP600URT) and coupled to a silicon-based avalanche photodetector (APD) (Thorlabs, APD430A2) with an active area diameter of 0.2 mm through a 100× objective lens, as well as a 450-nm long-pass filter (Thorlabs, FELH0450). The APD was then connected to a vector network analyzer. The vector network analyzer was pre-calibrated using an electronic calibration module (Agilent, 85093-60010). The DC input of the LD was fixed at 65 mA or 4.2 V with an output power of 45.7 mW. The peak-to-peak amplitude (V_{pp}) of the small signal was 100 mV.

2.4. Data transmission measurement

For OOK modulation, a 405-nm laser diode (Nichia, NDV4316) mounted in the TEC was connected to the transmitter module of a bit-error-rate tester (BERT) (Anritsu, ME522A) through a variable attenuator and linear amplifier (Mini-Circuits, ZHL-6A+) to control the amplitude. The optical output from the perovskite-polymer-fiber–based detector was guided through a silica-based fiber and other free-space optics to a silicon-based APD for optical-to-electrical (OE) conversion, similar to the experimental setup used for frequency bandwidth measurement. A linear amplifier (Mini-Circuits, ZHL-6A+) and a variable attenuator were connected to the APD to control the amplitude, and the electrical signal was demodulated by the receiver module of BERT (Anritsu, ME522A). Eye diagrams were captured using a digital communication analyzer (Agilent, 86100C). The V_{pp} of the input data at the LD was approximately 2 V. For the QAM-OFDM modulation scheme, the data input was sent through an arbitrary waveform generator (AWG) (Tektronix, AWG70002A) and demodulated using a mixed domain oscilloscope (Tektronix, MDO3104). All sent and received signals were processed offline using MATLAB.

3. Results and discussion

3.1. Physical and material characterization of CsPbBr₃ NCs

Figures 1(a) and (b) show TEM images of the CsPbBr₃ NCs acquired for the present study. Uniform cubic-shaped NCs were observed. The interplanar spacing (d-spacing) examined using high-resolution TEM images shown in Fig. 1(b) was determined to be ~0.29 nm, corresponding to the (002) crystal lattice planes of CsPbBr₃ [34]. The average edge length of the cubic-shaped NCs, as examined from the TEM images, was approximately 6.36 ± 0.03 nm (see Fig. 1(c)) and thus fell within the strong quantum confinement regime (i.e., <7 nm edge length) which was determined based on the dimensions of NCs relative to the exciton Bohr diameter [35]. The use of strong confinement NCs ensures high defect tolerance (e.g., through ligand passivation), size-tunability, and high quantum yield photoluminescence, with a narrow linewidth and fast transition lifetime suitable for high-speed data transmission/detection, which is lacking in most organic dyes [36,37]. Figure 1(d) shows the room-temperature XRD pattern of the CsPbBr₃ NCs, where distinct peaks were located at 20 values of 14.6°, 20.9°, 30.0°, 33.8°, and 37.3°, corresponding to the (100), (110), (200), (210), and (211) planes of CsPbBr₃. The crystalline structure of the CsPbBr₃ NCs resembles that of the cubic-dominated phase, rather than the orthorhombic phase, because of the lack of signature double peaks at the 20 value of 30° [38].
Fig. 1. Physical and material characterization of CsPbBr$_3$ NCs. (a) TEM and (b) HRTEM images of CsPbBr$_3$ perovskite NCs. (c) Corresponding edge length distribution histogram of the NCs. (d) X-ray diffraction (XRD) spectra of the CsPbBr$_3$ NCs compared to a standard spectrum extracted from the Inorganic Crystal Structure Database (ICDS). The inset depicts the cubic structure of the CsPbBr$_3$ crystal, where the lattice constants (a, b, c) are symmetrical.

3.2. Perovskite NCs-polymer scintillating fiber

Having established the physical and material properties of CsPbBr$_3$ NCs, the NCs were embedded into a polymer-based fiber, as shown in Fig. 2(a). The CsPbBr$_3$ NCs were dissolved in a UV-curable monomer, i.e., isobornyl acetate (IBOA), and a photoinitiator with varying concentrations from 20 mg/mL to 0.2 mg/mL for subsequent experiments. The cladding layer of the polymer-based fiber with an outer diameter of 4 mm was fabricated of polydimethylsiloxane (PDMS) with a refractive index of 1.436 using the molding method. Once the hollow-core fiber preform was obtained, the CsPbBr$_3$ NC-polymer solution filled the core with an inner diameter of approximately 937 $\mu$m through the injection molding method and was subsequently cured using UV irradiation ($\lambda_{\text{peak}} = 365$ nm) for 20 min. The CsPbBr$_3$ NCs, with a refractive index of approximately 1.9 [39], constituted less than 1% of the core layer based on the mole fraction. The refractive index of 1.476 was determined predominantly by the IBOA polymer (>99%) based on the conventional Arago-Biot equation [40], which is given by $n = \sum_i \phi_i n_i$, where $\phi_i$ is the mole fraction of the $i^{th}$ component. Although the core layer was optimized with varying NC concentrations, the refractive index of the core layer relies on the primary IBOA monomer as the luminescent dye constitutes only between 1% (20 mg/mL) and 0.01% (0.2 mg/mL) of the molecular ratio in the CsPbBr$_3$-IBOA mixture.
Fig. 2. (a) Cross-sectional micrograph images of the CsPbBr$_3$ NC-polymer fiber, where the CsPbBr$_3$ NCs-IBOA composite formed the core layer of the fiber, while the cladding layer was fabricated of PDMS. (b) Cross-sectional micrograph images for the core layer of the fiber formed by CsPbBr$_3$ NC-IBOA composite with a core diameter of $\sim$957 $\mu$m under optical excitation. The top diagram illustrates NCs embedded inside the IBOA polymer, and the re-emitted light propagates through to the fiber ends via total internal reflection when excited by a light source. Images of the CsPbBr$_3$ NCs-polymer fiber (c) without illumination (under ambient light) and (d) under UV illumination. Images of the (e) polymer-based fiber detector array formed by the single CsPbBr$_3$ NCs-polymer fiber and (f) when submerged in a water bath.

Figure 2(b) shows a microscopic image of the core layer consisting of the UV-cured CsPbBr$_3$ NC-polymer blend when illuminated by a 405-nm diode laser. The general concept of the perovskite-polymer scintillating fiber is also illustrated in Fig. 2(b), where NCs embedded inside the polymer matrix were excited by an incoming light source through the cladding layer and re-emitted from the polymer-fiber ends via total internal reflection (TIR) owing to the change in the refractive index. Based on Snell’s law, the critical angle for achieving TIR in the perovskite-polymer fibers was estimated to be approximately 76.63$^\circ$ by considering the difference between the refractive indexes of the cladding ($n = 1.436$) and core ($n = 1.476$) layers. While modifying the refractive indices of the core and cladding layer remained as a more straightforward method in optimizing the trapped light power and output power coupled from the fibers’ facet [41,42], increasing the diameter of the core layer could also ensure that more incoming light could be easily captured as compared to a smaller diameter which may significantly reduce the signal.
output power. At the same time, the diameter of the cladding layer should also be further reduced in the future in order to increase the light detection area that is primarily determined by the diameter of the core layer especially when arranged in an array form. Images of the as-fabricated perovskite-polymer fiber are shown under ambient lighting conditions and UV illumination, respectively, in Figs. 2(c) and (d). The inset of Fig. 2(d) further shows the cross-sectional image of the perovskite-polymer fiber when illuminated by a UV light source, where the light emission from the CsPbBr$_3$ NCs doped inside the core layer could be observed. For a proof-of-concept demonstration, the maximum length of the fabricated fiber was restricted to 30 mm, owing to the limitation of the plastic mold used in our work. However, it is envisaged that similar techniques of fiber drawing used in conventional glass-based fiber manufacturing could be employed in the future for high-volume manufacturing and scalability, following the development of perovskite crystal preforms that can sustain the high processing temperatures employed for the fiber-drawing technique. The as-fabricated single-perovskite-polymer fiber could be used to form an array of polymer-based fiber detectors, as shown in Fig. 2(e) under UV illumination. The facile method of forming a large-area detector by cascading or extending, either by the length or number of scintillating fibers, allows flexibility in scaling up the photodetection area without being limited by the conventional RC limit in planar detectors. The perovskite-polymer-based fiber detector array was also tested by submerging it in a water bath filled with deionized water, as shown in Fig. 2(f), thus highlighting its potential for practical deployment in underwater wireless optical communication systems [5].

Meanwhile, to maximize the optical gain at the receiver end while employing the scintillating fibers containing luminescent particles for the aforementioned photodetection purpose, the potential reabsorption losses introduced by the luminescent particles embedded within the light guiding medium must be reduced. The ideal condition would be negligible or zero overlap between the absorption and emission spectra. However, the highly defect-tolerant CsPbBr$_3$ perovskite NCs exhibit a size-dependent Stokes shift of approximately 82 to 20 meV [43]; thus, the reabsorption losses originating from the embedded NCs cannot be neglected. Figure 3(a) shows the absorption and emission spectra of the CsPbBr$_3$ NC-polymer solution with varying concentrations from 20 mg/mL to 0.2 mg/mL. When the NC concentration was gradually reduced, the absorption edges exhibited a blue-shift behavior from 521.7 nm (20 mg/mL) to 511.6 nm (0.2 mg/mL) with the gradual appearance of a hump in the vicinity of 495 nm, observed at lower concentrations (i.e., ≤2 mg/mL). In contrast, the photoluminescence (PL) intensity reduced without significant shifting of the peak position (i.e., in the vicinity of 515 nm) observed. The evident appearance of the sharp hump in the absorption spectrum is typically assigned to excitonic transitions of colloidal nanocrystals [44,45]. The capping ligands may have introduced interfacial electronic states coupled to the NCs and shifted the optical band gap, such as those reported for cadmium selenide (CdSe) quantum dots [46], whereas reducing the NC concentration reduced these capping ligands and exposed the inherent electronic states of NCs. By comparing the overlapped area under the curve of the emission and absorption spectra, the overlapping percentage substantially reduces from 57.7% (20 mg/mL) to 17.3% (0.2 mg/mL) (see Supplement 1 Fig. S1). Nevertheless, notably, the PL intensity also significantly reduces owing to the reduction in the NC concentration and may hamper the received optical power when used for photodetection. Therefore, given the fact that the absorption edge (i.e., 514.2 nm) does not overlap with the peak emission wavelength (i.e., 514.9 nm), the NC concentration of 2 mg/mL was employed in subsequent experiments because of its relatively higher emission intensity (i.e., more than a factor of 2 as compared to that of lower concentrations) and a reduced overlapping percentage as compared to that at higher concentrations.

Figure 3(b) shows the absorption spectra of the CsPbBr$_3$ NC-IBOA composite at the chosen NC concentration of 2 mg/mL and those of pristine IBOA, along with the corresponding PL emission spectra of the CsPbBr$_3$ NC-IBOA mixture. The CsPbBr$_3$ NC-IBOA composite exhibited strong
Fig. 3. Optical characterization of the CsPbBr$_3$ NCs-IBOA polymer fiber. (a) Absorption and photoluminescence (PL) spectra of the CsPbBr$_3$ NC-IBOA composite with varying concentrations from 20 mg/mL to 0.2 mg/mL. (b) Comparison of the absorption spectra of pristine IBOA with those of the CsPbBr$_3$ NC-IBOA composite, as well as the PL spectrum of the CsPbBr$_3$ NC-IBOA composite with the peak emission located at 514.9 nm. (c) Photostability of the CsPbBr$_3$ NC-IBOA composite and pristine CsPbBr$_3$ NCs under 405-nm laser excitation over a period of >30 h. (d) Time-resolved photoluminescence (TRPL) decay trace of the CsPbBr$_3$ NCs-IBOA composite and pristine CsPbBr$_3$ NCs under 425-nm excitation.

absorption beyond the band edge and toward the shorter wavelength region. However, notably, the cured IBOA used in this work showed an increased absorption toward the shorter wavelength region, i.e., <400 nm, which signifies that the absorption at this region is primarily dominated by IBOA and would lead to a lower quantum conversion efficiency. In such cases, the ideal incident wavelength for the CsPbBr$_3$-IBOA polymer-based fibers would lie in the wavelength region of approximately 400 to 475 nm, where the absorption effect from IBOA is minimized. The photoluminescence excitation (PLE) spectra of the CsPbBr$_3$-IBOA composite layer, as shown in Fig. S2, also revealed significant roll-off in terms of PL intensity for excitation wavelengths below 400 nm. The PL and absorption spectra of the as-synthesized CsPbBr$_3$ are also included in Fig. S3, where the peak emission is located at 509.5 nm with a full-width at half-maximum (FWHM) of 19 nm. This peak resembles that of the CsPbBr$_3$-IBOA composite shown in Fig. 3(b), except that the addition of UV-curable monomer (i.e., IBOA and PI) ensures that the luminescent dye, i.e., CsPbBr$_3$ NCs, can be embedded and fully cured inside the core layer of polymer-based fibers. A comparison of photostability of the CsPbBr$_3$ NCs and CsPbBr$_3$ NC-IBOA under excitation with a 405-nm laser diode is also shown in Fig. 3(c). Clearly, the PL intensity of pristine CsPbBr$_3$ NCs deteriorates substantially over the measurement period of >30 h as compared to that of the CsPbBr$_3$ NC-IBOA composite, where the polymer composite provided an additional matrix to minimize exposure of the NCs to the ambient environment and reduced structural variation.
(e.g. strain disorders and ion migration) [47]. An image of the as-prepared CsPbBr\(_3\) NC-IBOA composite solution is shown in Fig. S4(a), revealing that the NCs can be dissolved completely in the IBOA monomer without any clear partitioning. Further characterization based on the Raman spectra of the CsPbBr\(_3\) NC-IBOA composite under 532-nm and 633-nm excitation (see Supplement 1 Fig. S4(b)) also revealed signature peaks (i.e., ~87, 128, and 309 cm\(^{-1}\)) which originated from the vibrational mode of the [PbBr\(_6\)]\(^4-\) octahedron [48] and distinct peaks (i.e., ~487, 625, and 632 cm\(^{-1}\)) from pristine IBOA.

To evaluate the feasibility of the proposed perovskite NCs as luminescent materials for wavelength conversion and optical detection, the time-resolved photoluminescence (TRPL) of the CsPbBr\(_3\)-IBOA composite was also characterized. Figure 3(d) shows the PL decay trace of both pristine CsPbBr\(_3\) and the CsPbBr\(_3\) NC-IBOA composite monitored at their respective peak wavelengths, which exhibits a radiative recombination lifetime (\(\tau\)) of 4.66 ns and 2.96 ns, respectively, fitted based on a single exponential function. To enable high-speed optical detection, the theoretical \(f_{\text{-3dB}}\) can be estimated using Eq. (1), as follows:

\[
f_{\text{-3dB}} \leq \frac{1}{2\pi\tau}
\]  

Accordingly, the theoretical maximum frequency bandwidth of the CsPbBr\(_3\) NC-IBOA composite is approximately 53.77 MHz. A comparison of the pristine and polymer composite samples, as well as the polymer composites of varying concentrations (see Supplement 1 Fig. S5) showed that no direct correlation was observed as a result of the polymer mixture or NC concentration, which leads to substantial variation in terms of the radiative recombination lifetime of the NCs. The resulting radiative recombination lifetime of the CsPbBr\(_3\) NC-polymer composite is comparable to that of other polymer-based luminescent materials used for similar wavelength-converting optical detection applications, for example, SuperYellow [21], Cm6 [18] and DCM [18], with a reported lifetime of less than 3 ns, which is substantial for supporting high-bandwidth OWC systems. The PLQYs of both samples, that is, pristine CsPbBr\(_3\) NCs and the CsPbBr\(_3\) NC-IBOA composite, were measured to be \(\geq 95\%\), which is higher than that of other reported polymer-based dyes, for example, SuperYellow (60%) [21], Cm6 (80 to 95%) [16,18] and DCM (50%) [18].

### 3.3. Optical modulation based on NRZ-OOK

To further elucidate the feasibility of the perovskite NC polymer fiber-based detector for OWC links, the experimental setup shown in Fig. 4(a) was used. A 405-nm laser diode (LD) was employed as the data transmitter, and its output light propagated through a free-space channel before being detected by the proposed perovskite-polymer fibers. A 450-mm-long silica-based fiber with a core diameter of 600 \(\mu\)m was used to couple the down-converted light from the scintillating fiber and guide it through free-space optics, i.e., a 450-nm long-pass filter and focusing lenses, before focusing on a compact silicon-based APD with an active area diameter of 0.2 mm and \(f_{\text{-3dB}}\) of 400 MHz. As previously mentioned, when the fabrication of the perovskite-polymer fibers was scaled up by, for example, using fiber-pulling towers, the output light could be directly coupled from the scintillating fiber onto the APD without the need for silica-based fibers, as demonstrated previously [19,20]. Removing the silica-based fiber could also reduce the coupling loss between the NCs-polymer fiber and silica-based fiber, as well as the propagation loss (~28 dB/km) that arises when transmitting the re-emitted light in the vicinity of 514 nm. The inset of Fig. 4(a) shows the down-converted light output from the silica-based fiber when the perovskite-polymer fiber was excited by a 405-nm laser diode through the cladding layer. Prior to evaluating the small-signal modulation of the fiber-based detector, the angle of view (AoV) of a single perovskite-polymer fiber with respect to its polar (\(\theta\)) and azimuthal (\(\phi\)) angles is also shown in the polar plots of Figs. 4(b) and (c), respectively, where the received...
optical power remained relatively uniform in the vicinity of -35 dBm throughout the angles of measurement, thus highlighting its potential to form a near-omnidirectional detector. Compared to an étendue-limited commercial planar detector with a typical AoV of up to 120° (or semiangles of 60°) (see Supplement 1 Fig. S6), despite exhibiting a lower optical gain that can be enhanced by stacking more fibers and increasing their length (limited by the critical length) and through the back-end electrical amplifiers, the advantages of the perovskite-polymer fiber in offering near-omnidirectional optical detection could be the primary choice for a dynamic OWC link. Thus, it can mimic the present radio-frequency antennas, where data signals can propagate from all directions.

By sweeping the sinusoidal AC modulation signal from 300 kHz to 200 MHz, as shown in Fig. 4(c), the $f_{-3dB}$ of the perovskite NC-polymer fiber was determined to be approximately 13.10 ± 0.47 MHz when measured across different lengths from the edge of the sample with a covering slit. As the $f_{-3dB}$ remained consistent irrespective of the illuminated length from the output edge, the effect of pulse spreading did not affect the overall modulation response of the sample. The critical length ($L_c$) of the scintillating fiber, where the effect of pulse spreading becomes the dominant factor for the frequency response, can be estimated as discussed by Peyronel et al. as follows [19]:

$$L_c \approx \frac{3.791\tau c}{n_{co}}$$  

(2)

where $\tau$ is the exponential decay lifetime of the luminescent particle, $c$ is the speed of light in vacuum, and $n_{co}$ is the refractive index of the core layer. Accordingly, $L_c$ can be estimated to be approximately 2.28 m for our sample and therefore affirms the scalability to further expand into a larger detection area (increasing the geometrical optical gain) without sacrificing the modulation bandwidth, as opposed to the case of planar detectors. The inset of Fig. 4(d) also shows the received optical power (i.e., -29.5 to -34.4 dBm) coupled with the silicon-based APD when the perovskite-polymer fiber sample was excited by the 405-nm LD with an excitation power of 45.7 mW. Subsequently, using the collimated 405-nm LD as the excitation source to scan across different sections of the perovskite-polymer fiber, the self-absorption loss can be determined using the expression below governed by Beer-Lambert law [21]:

$$I = I_0 e^{-\alpha x}$$  

(3)

where $I$ denotes the received optical power across different sections of the polymer fiber, $I_0$ is the received optical power at the edge of the sample, assuming negligible self-absorption loss, $\alpha$ is the self-absorption loss, and $x$ is the distance measured from the fiber edge. Based on the governing equation above, the self-absorption loss was estimated to be 0.0615 cm$^{-1}$ (see Supplement 1 Fig. S7(a)), which is much lower than that of a previously demonstrated polymer-based luminescent concentrator (0.5 cm$^{-1}$) and with a much narrower AoV (±55°) [21]. Furthermore, to demonstrate the communication performance of the perovskite-polymer fiber detector, that is, using the experimental setup shown in Fig. 4(a), a gross data rate of 23 Mbit/s with a bit error ratio (BER) of $2.3 \times 10^{-3}$, below the 7% overhead forward error correction (FEC) limit of $3.8 \times 10^{-3}$, was achieved based on the non-return-to-zero on-off keying (NRZ-OOK) modulation scheme with a pseudorandom binary sequence (PRBS) $2^{15}-1$ data format when the incoming optical beam was focused onto the furthest section of the fiber (i.e., 30 mm from the edge of the fiber), as shown in Fig. 4(e). The insets of Fig. 4(d) show the corresponding eye diagrams at 23 Mbit/s and 15 Mbit/s with a BER of $2.5 \times 10^{-7}$. Further measurements with a fixed modulation data rate of 23 Mbit/s conducted at different sections of the perovskite-polymer fiber detector up to a distance of 30 mm also confirmed that the BER remained below the FEC limit across the section of the fiber (see Supplement 1 Fig. S7(b)).
Fig. 4. Optical modulation measurement using the CsPbBr$_3$ NC-polymer fiber-based detector. (a) Experimental setup for data rate measurement using the non-return-to-zero on-off-keying (NRZ-OOK) modulation scheme. The inset shows the optical output from the silica-based fiber coupled with the CsPbBr$_3$ NC-polymer fiber, where the excitation light from the 405-nm laser diode is absorbed/detected by the perovskite-polymer fiber and down-converted to a longer wavelength in the vicinity of 514 nm. The measured received optical power from the CsPbBr$_3$ NC-polymer fiber with respect to its (b) polar ($\theta$) and (c) azimuthal ($\phi$) angles. (d) Normalized frequency response of the CsPbBr$_3$ NC-polymer fiber when measured at different lengths, i.e., 5 to 30 mm, from the edge of the sample. The inset shows the corresponding –3-dB frequency bandwidth ($f_{-3dB}$) and the received optical power at different sections of the fiber. (e) Bit error ratio (BER) of data transmission based on the NRZ-OOK modulation scheme at different data rates. The insets detail the corresponding eye diagrams at 23 Mbit/s with a BER of $2.3 \times 10^{-3}$ and at 15 Mbit/s with a BER of $2.5 \times 10^{-7}$. 
3.4. Optical modulation based on QAM-OFDM in UWOC

To further improve the spectral efficiency of the communication link and shed light on the viability of the proposed perovskite-polymer fiber for high-bit-rate detection in the UWOC link, an orthogonal frequency-division multiplexing (OFDM) scheme with adaptive bit- and power-loading, which is known to exhibit higher spectral efficiency as compared to OOK modulation formats, was subsequently implemented based on the experimental setup shown in Fig. 5(a). Owing to the much lower absorption coefficient ($<1\,\text{m}^{-1}$) of visible wavelengths in water as compared to that of acoustics and radio frequency communication, UWOC has emerged in recent years as a secure and high-bitrate communication modality to support various tactical underwater activities where connection via fiber optics is physically challenging, for example, submarine communication, environmental surveying via autonomous underwater vehicles (AUVs), and underwater sensor nodes [4]. Figure 5(b) shows photographs of the proof-of-concept experimental setup, where an intensity-modulated 405-nm laser diode was used for signal transmission across a 1-m–long water tank filled with the American Society for Testing and Materials (ASTM) Type 1 Reagent Grade water. On the other end, the perovskite-polymer fibers were completely submerged underwater for signal detection before being optically guided to a compact silicon-based APD.

In brief, the PRBS $2^9-1$ data format was generated and converted from serial to parallel before being modulated. Hermitian symmetry was then introduced to force the output of the inverse fast Fourier transform (IFFT) to be real-valued, followed by the addition of the cyclic prefix. The formed array of OFDM symbols can then be converted to a serial sequence and used for the intensity modulation of the transmitter. At the receiver end, the OFDM symbols were recorded using an oscilloscope and resampled down to the sampling frequency of the AWG. The signal was synchronized using known training symbols. Accordingly, the received signal was converted
from serial to parallel, and subsequently, the cyclic prefix was removed before performing FFT. After removing the Hermitian symmetry symbols, a single-tap equalizer based on the introduced training symbols was used for post-equalization, and the symbols were demodulated. The received bits can then be converted to a serial sequence, and the BER of the optical communication link can be calculated. In this demonstration, a 2-quadrature amplitude modulation (2-QAM) signal with uniform bit loading was first employed to estimate the signal-to-noise ratio (SNR) calculated based on the error vector magnitude (EVM), where the performance for each subcarrier can be obtained. Depending on the estimated SNR and channel capacity \( C \), the number of bits can be subsequently allocated, as shown in Fig. 5(c). The channel capacity determined from the Shannon limit is governed by Eq. (4), as follows:

\[
C = \log_2(1 + SNR) \tag{4}
\]

In this case, no bits were allocated to subcarriers with SNRs below 3 dB. Figure 5(d) shows the SNR and power-loading factor for each carrier. Based on the used frequency bandwidth of 1.2 to 50 MHz, the total number of subcarriers \( N_{SC} \) of 200, the FFT size \( N_{FFT} \) of 1024, as well as 150 OFDM symbols with a cyclic prefix length \( N_{CP} \) of 10, the gross data rate can be calculated using Eq. (5), as follows:

\[
D = \frac{f_{AWG}}{N_{FFT} + N_{CP}} \sum_{k=1}^{N_{SC}} \log_2(M_k) \tag{5}
\]

where \( f_{AWG} \) is the sampling frequency of the AWG, that is, 250 MSample/s, \( M_k = 2^b \) is the QAM order of the \( k^{th} \) subcarrier, and \( b \) is the number of allocated bits. A gross data rate of 175.3 Mbit/s was achieved with an average BER of 1.1 \times 10^{-3}, which is well below the required FEC limit of 3.8 \times 10^{-3}. The BER for each subcarrier is shown in Fig. 5(e). The superimposed constellations corresponding to 4-, 8-, and 16-QAM for subcarriers of the same order are included in the inset. Assuming a 7% FEC overhead and 6% training symbols used for synchronization and post equalization, the net data rate was estimated to be 152.5 Mbit/s. While the net data rate was achieved with a transmitted power of 45.7 mW, further characterizations in terms of the influence of transmitted power on the channel capacity revealed a gradual overall reduction with reduced transmitted power that will impose a trade-off with the data rate (see Supplement 1 Fig. S8(a)). The normalized channel capacity, as measured using the area under the curve of the channel capacity versus subcarrier index (see Supplement 1 Fig. S8(b)), can be used to estimate the net data rate for cases in which the transmitted power must be reduced for energy-saving purpose when operating in a power-hungry environment. A 4-QAM signal with a net data rate of 87.4 Mbit/s transmitted across the channel revealed that the BER remained below the FEC limit for up to ~94% reduction (or equivalent to approximately 2.7 mW) in the transmitted power (see Supplement 1 Fig. S8(c)). This may also signify that the transmission distance could be further increased but that requires significant experimental validation in the future that falls out of the scope of the present work. The demonstrated data rate in the range of hundreds of Mbit/s is sufficient to support various short-haul tactical activities in underwater environments, when compared with conventional communication modalities based on acoustics or radio-frequency signals that can only support data rates of up to kbit/s, and the use of the scintillating fiber-based receiver also addresses the PAT issues when scaling up to meter-long detection sizes in the future. Before employing into a practical underwater environment, further studies and evaluation of the BER performance of the NCs-polymer fibers-based receiver due to water turbulence (e.g., air-bubble-, temperature-, salinity-, turbidity-, and mobility-induced) would also be beneficial in order to ensure the reliability of a UWOC system [49].

Table 1 summarizes various types of optical detectors recently conceived for optical communication links. Compared with conventional planar-based detectors, where the detection area suffers from a trade-off with the modulation bandwidth owing to the RC-limitation [31,32,50,51],
luminescent concentrator/fiber-based detectors overcome this limitation as the detection area is scalable, for example, by arranging in a fiber array form [19], because the frequency bandwidth is governed by the carrier lifetime of the luminescent particles. This methodology offers a scalable, large-area and near-omnidirectional optical detection module that could relieve the alignment issues especially when operating in a harsh environment. Moreover, as compared to recently demonstrated fiber-optic tapers for increasing the detection area [52], albeit having a lower modulation bandwidth, these fiber-optic taper structures are etendue-limited, are rigid in nature, and are non-omnidirectional. Moreover, they require multi-pixel detectors to maintain a high optical gain, a wide field of view, and a broad modulation bandwidth. The optical gain (\( G \)) is defined by Eq. (6) [19,21]:

\[
G = \frac{A_{\text{input}}}{A_{\text{output}}} \eta_{\text{abs}} \eta_{\text{PLQY}} \eta_{\text{prop}} \eta_{\text{col}}
\]

(6)

where \( A_{\text{input}} \) is defined as the active area for light absorption, \( A_{\text{output}} \) is the active area of the detector coupled to the luminescent fiber/concentrator, \( \eta_{\text{abs}} \) is the light efficiency absorbed by the luminescent detector, \( \eta_{\text{PLQY}} \) is the photoluminescence quantum efficiency of the luminescent particles, \( \eta_{\text{prop}} \) is the propagation efficiency within the waveguide medium, and \( \eta_{\text{col}} \) is the collection efficiency due to coupling to the detector. The achieved optical gain could be indefinitely large because the input area can be expandable (i.e., by using either arrays of multiple fibers or multiple rounds of bending based on a single fiber below the critical length) to a few orders of magnitude larger than the output receiver end without sacrificing the modulation bandwidth, despite several loss mechanisms originating from surface absorption, fiber coupling, re-absorption, and light conversion losses. Based on the measurement of a single perovskite-polymer fiber, the optical gain is expected to exceed 2 by scaling to a closely packed detector array based on tens or more scintillating fibers, which provides a sufficiently large detection area similar to the solar cell-based receiver while maintaining a non-RC-limited modulation bandwidth. However, as highlighted in Eq. (2), the maximum length of the luminescent fibers may need to be controlled well below the critical length where the effect of pulse spreading dominates the modulation bandwidth with a steeper roll-off at higher frequencies [19]. Similarly, while arranging the luminescent fibers in an array configuration or when illuminating multiple closely packed luminescent fibers with a light source with a wider divergence angle (e.g., LEDs), the effect of timing jitter may be significant due to the inhomogeneous distribution of luminescent particles doped within the fiber, leading to variations in the modulation bandwidth. Such constraints in terms of uniformity need to be resolved when scaling up the technology or deploying into a practical system. In contrast, the frequency bandwidth of perovskite nanocrystals demonstrated herein is still lacking compared to those of other organic-based luminescent materials. However, owing to the wide wavelength tunability of halide perovskite-based nanocrystals, spanning across the visible-to-near-infrared region simply by tuning the constituent composition, it could also be anticipated that wavelength-division multiplexing (WDM) could be realized based on scintillating fiber arrays cascaded based on perovskite-polymer-based fibers of different emission wavelengths, for example, I- or Cl-based perovskites, for supporting various optical-based communication systems requiring higher bitrates and channel capacities. Moreover, the higher photoluminescence quantum yield of the perovskite nanocrystals allows a higher signal-to-noise ratio, which can improve the reliability of the link and the data rate.
Table 1. Comparison of various types of optical detectors for optical wireless communication.

<table>
<thead>
<tr>
<th>Type of optical detector</th>
<th>Material</th>
<th>PLQY</th>
<th>Operating Wavelength</th>
<th>Channel</th>
<th>$f_{-3dB}$ bandwidth</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Planar</td>
<td>CsPbBr$_2$, CsPbBr$_3$</td>
<td>-</td>
<td>450 nm</td>
<td>FSO</td>
<td>0.6 MHz, 1 MHz (RC-limited)</td>
<td>Bao et al. [31]</td>
</tr>
<tr>
<td>Planar</td>
<td>Cs$<em>x$FA$</em>{1-x}$Pb(I$_{1-y}$Br$_y$)</td>
<td>-</td>
<td>540 nm</td>
<td>FSO</td>
<td>120 kHz (RC-limited)</td>
<td>López-Fraguas et al. [32]</td>
</tr>
<tr>
<td>Planar</td>
<td>PTB7:PC71BM, PTB7-Th:PC71BM, PTB7-Th:EH-IDTBR</td>
<td>-</td>
<td>660 nm</td>
<td>FSO</td>
<td>1.32 MHz, 1.26 MHz, 2.77 MHz (RC-limited)</td>
<td>Tavakkolnia et al. [50]</td>
</tr>
<tr>
<td>Solar cell</td>
<td>a-Si</td>
<td>-</td>
<td>Broadband White Light</td>
<td>FSO &amp; UWOC</td>
<td>290 kHz (RC-limited)</td>
<td>Kong et al. [51]</td>
</tr>
<tr>
<td>Organic polymer-luminescent concentrator</td>
<td>SuperYellow</td>
<td>~60%</td>
<td>450 nm</td>
<td>FSO</td>
<td>100 MHz (carrier lifetime-limited)</td>
<td>Dong et al. [21]</td>
</tr>
<tr>
<td>Organic polymer-luminescent fiber</td>
<td>Organic dye</td>
<td>&lt;50%</td>
<td>405 nm</td>
<td>FSO</td>
<td>91 MHz (carrier lifetime-limited)</td>
<td>Peyronel et al. [19]</td>
</tr>
<tr>
<td>Inorganic perovskite-luminescent fiber</td>
<td>CsPbBr$_3$</td>
<td>≥95%</td>
<td>405 nm</td>
<td>FSO &amp; UWOC</td>
<td>13.10 MHz (carrier lifetime-limited)</td>
<td>Present work</td>
</tr>
</tbody>
</table>

4. Conclusions

We fabricated proof-of-concept CsPbBr$_3$ nanocrystal-polymer-based scintillating fibers and demonstrated their application as a near-omnidirectional detector for a visible-light-based optical communication link. The proposed perovskite NC-polymer-based fibers omit the conventional RC limitation, as demonstrated in planar-based optical detectors, and provide a scalable large detection area with an $f_{-3dB}$ of 13.10 MHz. We further demonstrated that data rates of up to 23 Mbit/s and 152.5 Mbit/s can be achieved based on NRZ-OOK and $M$-QAM OFDM modulation schemes, respectively. Along with further extensive efforts in terms of high-volume manufacturing (i.e., based on the fiber pulling method) and reducing the carrier recombination lifetime to the sub-nanosecond range, we anticipate that the wavelength-tunable perovskite NC-polymer-based fibers could be a strong candidate for future omnidirectional transceiver units in high-bit-rate terrestrial- and underwater-internet systems.

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Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

Supplemental document. See Supplement 1 for supporting content.
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