Perovskite-Quantum-Wires-Based Full-Color Fiber Light-Emitting Diodes for Flexible Electronics

Beitao Ren[1,2], Zhiyong Fan[1,2]

[1]Department of Electronic and Computer Engineering, HKUST, Kowloon, Hong Kong SAR, China

[2]State Key Laboratory of Advanced Displays and Optoelectronics Technologies, HKUST, Kowloon, Hong Kong SAR, China

Abstract

Fiber light-emitting diode (Fi-LED), which can be used for wearable lighting and display devices, is one of the key components for fiber/textile electronics. However, it remains impediments to overcome on device fabrication with fiber-like substrates, such as gravity-induced nonuniform coating, low-quality crystallization, etc. Here, we uniformly grew perovskite quantum wires (PeQWs) by filling high-density alumina nanopores on aluminum fibers by dip-coating. The obtained PeQWs exhibit a prominent photoluminescence quantum yield of nearly 90% and a noticeable photoluminescence lifetime of up to 1,500 hours under ambient condition. With further conformal deposition of transporting layer and electrode, as well as the polydimethylsiloxane encapsulation, full-color PeQWs-based Fi-LEDs with mechanical bendability, stretchability, and waterproof feature have been successfully demonstrated.

Author Keywords

Fiber/textile electronics; Wearable lighting/display; Perovskite quantum wires; Full-color fiber LEDs.

1. Introduction

Metal halide perovskites (MHPs) have emerged as highly promising emissive materials for the next-generation of light-emitting diodes (LEDs) due to their exceptional optoelectronic properties, including efficient radiative carrier recombination, impressive color tunability and purity, and straightforward synthesis methods at low temperatures in both liquid and vapor phases (1-4). Over the past decade, the external quantum efficiency (EQE) of perovskite LEDs (PeLEDs) has been increased dramatically from under 1% to over 30% (5, 6). Meanwhile, MHPs have also been successfully incorporated into upcoming technologies, such as large-scale, flexible, and multifunctional LEDs (7-10), indicating their significant potential in wearable display applications.

Fiber light-emitting diode (Fi-LED), which emits light from a flexible substrate resembling a fiber, offers distinct advantages in wearable displays due to its seamless integration with textile fabrication as well as excellent spatial luminance uniformity (11). However, challenges arise during the deposition of MHP thin films, as the solution coating on fiber-like substrates may lack uniformity because of gravitational and surface tension effects (8, 9). In this work, we demonstrate a dip-coating method for the uniform growth of perovskite quantum wires (PeQWs) on aluminum (Al) fibers, with the assistance of porous alumina membrane (PAM) templates. By harnessing the quantum confinement effect alongside passivation provided by the three-dimensional (3D) PAM structure, the obtained high-quality PeQWs exhibit a remarkable photoluminescence quantum yield (PLQY) approaching 90% and an extended photoluminescence (PL) lifetime (T_{PL50}) of up to 1500 hours even under ambient conditions. Through a two-step evaporation method to coat a surrounding transporting layer and semi-transparent electrode, we have successfully fabricated full-color Fi-LEDs. Intriguingly, additional polydimethylsiloxane (PDMS) encapsulation helps instill the mechanical bendability, stretchability, and waterproof characteristics of Fi-LEDs. The plasticity of Al fiber also allows the one-dimensional (1D) architecture Fi-LED to be shaped and constructed for two-dimensional (2D) or even 3D architectures, ushering in new possibilities for advanced flexible lighting with unconventional formfactors.

2. Results

High Quality Crystallized PeQWs

As shown in Figure 1a, MHP precursor [$CsPbX_3$ (X = Cl, Br, I, or mixed halide)] solution is filled into the PAM channels on the surface of a thin Al wire. This process results in an almost complete filling of the channels with a clean surface, as evidenced by a comparison of the top-view scanning electron microscopy (SEM) images of PAM before (Figure 1a2) and after (Figure 1a3) PeQWs growth. A well-crystallized PeQW extracted from PAM channel is characterized by high-resolution transmission electron microscopy (HRTEM) technology (Figure 1a4), revealing an interplanar spacing of 0.29 nm corresponding to the distance between two neighboring (200) planes of orthorhombic $CsPbBr_3$ phase. In Figure 1b, the SEM top-view images illustrate the morphological distinctions between $CsPbBr_3$ QWs and $CsPbBr_3$ thin film. Evidently, $CsPbBr_3$ QWs provide more uniform and brighter luminescence under ultraviolet (UV) stimulus (insets in

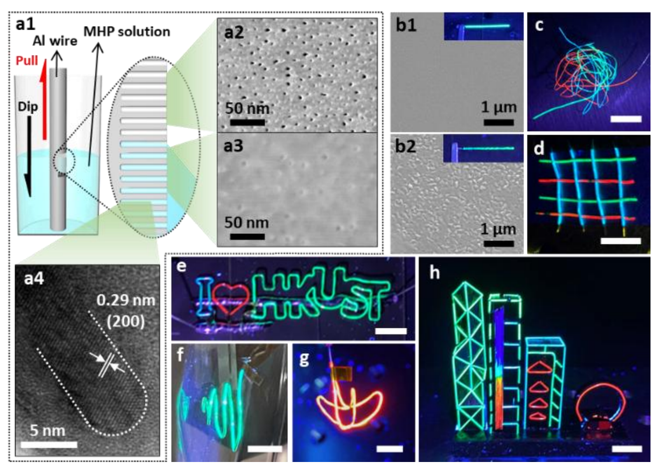

Figure 1. (a) Schematic of PeQWs formation by dip-coating process (a1) and the corresponding electron microscopy images: the SEM images of PAM before (a2) and after (a3) PeQWs formation, and the HRTEM image (a4) of a single $CsPbBr_3$ QW. (b) Comparison between PeQWs and MHP thin film. (c-h) Fluorescent pictures: tangled fibers (c), woven fibers (d), "I Love HKUST" pattern (e), green swirl (f), HKUST Redbird logo (g), and Victoria Harbor (h) built by mounting batches of fibers. Unlabeled scale bars: 1 cm.

ISSN 0097-996X/25/5601-00162-$1.00 © 2025

Figure 1b), which can be attributed to quantum confinement effect and the mitigation of surface tension during dip-coating. In addition, benefiting from the malleability of Al, the fibers can be tangled (Figure 1c) and woven (Figure 1d) with each other. Meanwhile, Al fiber has both malleability and plasticity, which allows it to be fashioned into diverse 2D and 3D architectures. Here, we showcase a 2D full-color string displaying "I Love HKUST" (Figure 1e), a 3D green swirl (Figure 1f), and an HKUST Redbird pattern (Figure 1g) with excellent fluorescence uniformity. In Figure 1h, we combine various types of PeQWs to build up a "night scene" of Victoria Harbour, where a halide exchange method is used to generate halide gradient for achieving color transition regions.

Figure 2a indicates the PL peak position and the PLQY of three representative red/green/sky-blue (R/G/B) PeQWs. Unlike their traditional thin film counterparts, PeQWs exhibit blue-shifted PL peaks located at 625 nm, 512 nm, and 490 nm respectively for R-, G-, and B- PeQWs, owing to the quantum confinement effect. The combination of spatial confinement and surface passivation facilitated by the PAM template contribute to high PLQY values of 59.96%, 87.11%, and 65.10%, respectively. Furthermore, the uniform growth of PeQWs on the PAM@Al fiber is confirmed through position-dependent PL measurements conducted along a sample exceeding 4 cm in length (Figure 2b). To further verify the protection provided by PAM template, the stability of PeQWs is quantitatively evaluated by monitoring the integrated PL intensity decay over time (Figure 2c). The as-grown PeQWs exhibit an extraordinary PL lifetime under ambient condition (23°C, 45% to 55% relative humidity), with the T_{PL50} values of ~720, ~1400, and ~1000 hours, respectively, for R-, G-, and B- PeQWs, where T_{PL50} represents the duration at which the PL diminishes to 50% of its initial intensity. Such remarkable stability in air is attributed to the PAM's ability to shield against moisture and oxygen.

Full-color flexible Fi-LEDs

To fabricate Fi-LED devices, a two-step evaporation method is employed to deposit a surrounding hole transporting layer (HTL) and a top transparent electrode (TE) onto the PeQWs. Figure 3a presents the schematic of our Fi-LED device, with copper phthalocyanine (CuPc) acting as the HTL and a thin layer of gold (Au) serving as the TE. The insets show the cross-sectional schematic of the Fi-LED and its corresponding SEM image, indicating the utilization of an approximately 200-nm-thick PeQW array structure. Note that an ultrathin Al_2O_3 layer between PeQWs and Al works as a tunnelling barrier for electron injection (12). When getting a high enough voltage bias for tunneling through the ultrathin Al_2O_3 layer, electrons engage in radiative recombination with holes within the PeQW region for light emission. For a convenient experimental estimation of Fi-LEDs performance, planar devices with identical structure and similar fabrication processes are characterized. The corresponding EQE-current efficiency-voltage (EQE-CE-V) curves are depicted in Figure 3b , showcasing peak EQE values of 7.6%, 11.6%, and 4.3% for R-, G-, and B- devices, respectively, at luminance levels of 144, 876, and 110 cd m^{-2}. Considering the severe electron

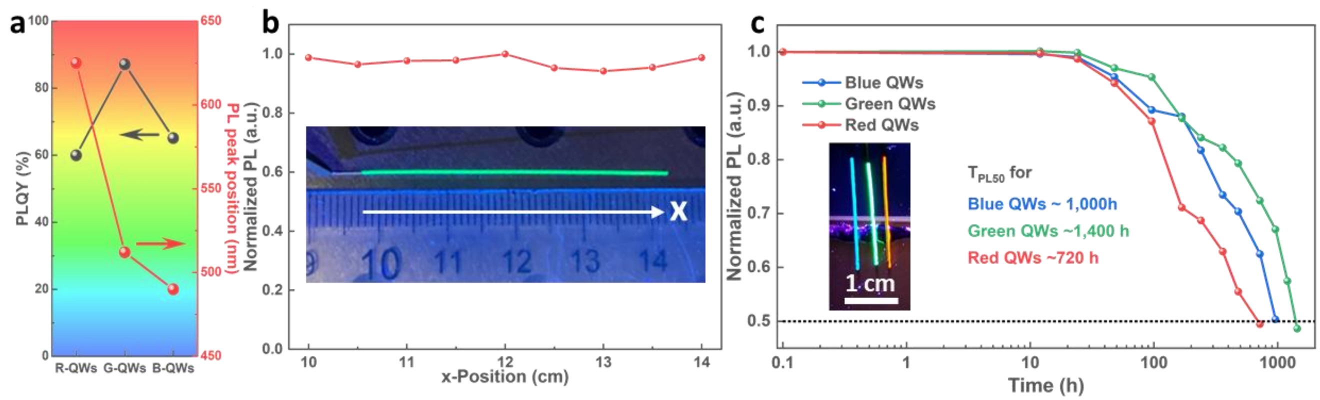

Figure 2. (a) PLQY and PL peak position of PeQWs with different halide compositions. (b) Normalized PL intensity evolution as a function of length position along a single fiber. The inset shows the fluorescent photograph of the corresponding green fiber. (c) PL stability test in ambient air (23℃, ~45-55% relative humidity), The inset is the fluorescent image.

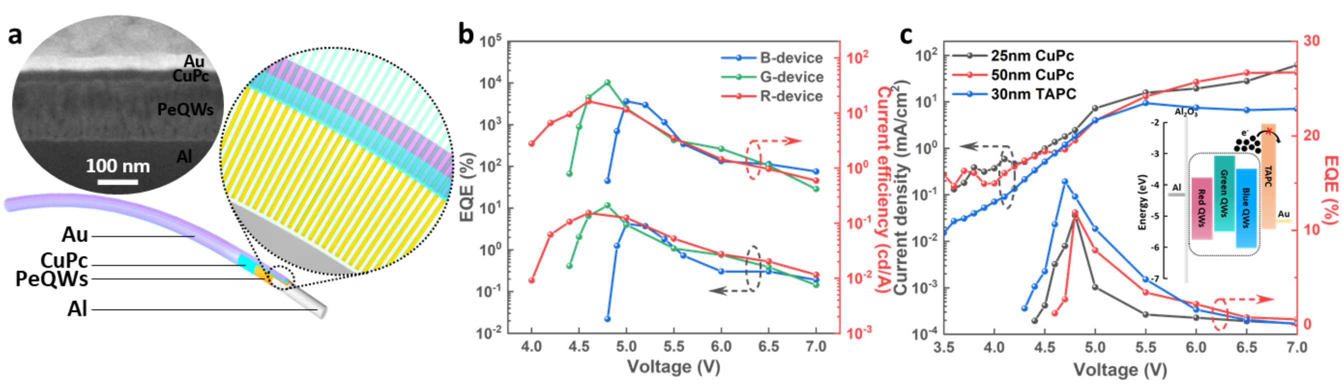

Figure 3. (a) Device schematic of Fi-LED. (b) EQE-CE-V curves of R/G/B PeLED devices. (c) Device characterizations with different HTLs. The inset indicates the modified energy band structure of TAPC devices for better electron blocking.

leakage from PeQWs because of the low LUMO (lowest unoccupied molecular orbital) of CuPc, further optimizations are conducted by increasing the thickness of CuPc and altering HTL from CuPc to 1,1-bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC). As demonstrated in Figure 3c, a thicker CuPc layer does not yield significant improvement, while the higher LUMO of TAPC substantially enhances the blocking effect for suppressing the electron leakage from PeQWs. This enhancement results in a notable increase in EQE, with the green PeQW-based device achieving an EQE of 15.2% (measured in ambient air condition without any encapsulation), which is close to 200 times higher than the previously reported PeLED devices with such a metal-insulator-semiconductor (MIS) structure (12).

Beyond their promising performance, the PeQW-based Fi-LEDs offer the capability to achieve light emission in both 2D and 3D configurations because Al fiber substrate can be reshaped into diverse profiles with 2D or 3D architectures, benefiting from both of its malleability and plasticity. As shown in Figure 4a, three Al fibers are sculpted to establish an "I Love HKUST" model, each tailored for B-, R-, and G- PeQW-based Fi-LEDs. The image captures the electroluminescent (EL) display under a constant 8-V bias. Notably, the "HKUST" string is crafted from a single 24-cm-long Al fiber, which, to the best of our knowledge, represents the longest monolithically independent perovskite Fi-LED documented in existing literature, with an emission area of nearly 4 cm^2. By encapsulating our Fi-LEDs with PDMS for resisting moisture and oxygen, as well as for stress dispersion, we have successfully developed PeQW-based Fi-LED devices with outstanding stretchability, bendability, twistability, and waterproof characteristics. Particularly, as illustrated in Figure. 4b, a wavy-shaped design, commonly utilized in stretchable and wearable electronics (13), has been implemented in our Fi-LED device to enhance its stretchability, achieving a maximum 100% elongation. Additionally, our Fi-LED maintains excellent EL performance with a bending radius of 3 mm (Figure 4c), or even a bending radius of 2 mm when employing a wavy structure device (Figure 4d). It is also interesting that the device remains undamaged even after undergoing 180° twisting (Figure 4e) or water soaking treatment (Figure 4f).

To further evaluate the robustness of Fi-LEDs with PDMS encapsulation, we monitored the EL degradation under various harsh conditions, including stretching, bending, and water soaking. In Figure 4g, we observed that the device sustains 85% of its initial value even under 100% strain (black curve), while it plummets to 50% after 150 stretching cycles at a 60% strain (red curve), indicating that the mechanical damage from the repeated stretching cycles is non-negligible. Figure 4h illustrates a 1.5-cm-long Fi-LED device attached onto cylinders with different diameters for bendability test. The EL intensity maintains approximately 80% of its original value when subjected to a bending radius of 2 mm (black curve). Meanwhile, after undergoing 500 bending cycles with a radius of 5 mm, the device preserves over 85% of its initial EL intensity (red curve). To improve device mechanical stability, thinner Fi-LED with a fiber diameter of 0.2 mm has been fabricated and measured. As a result, the device can maintain 80% of its EL intensity after 150

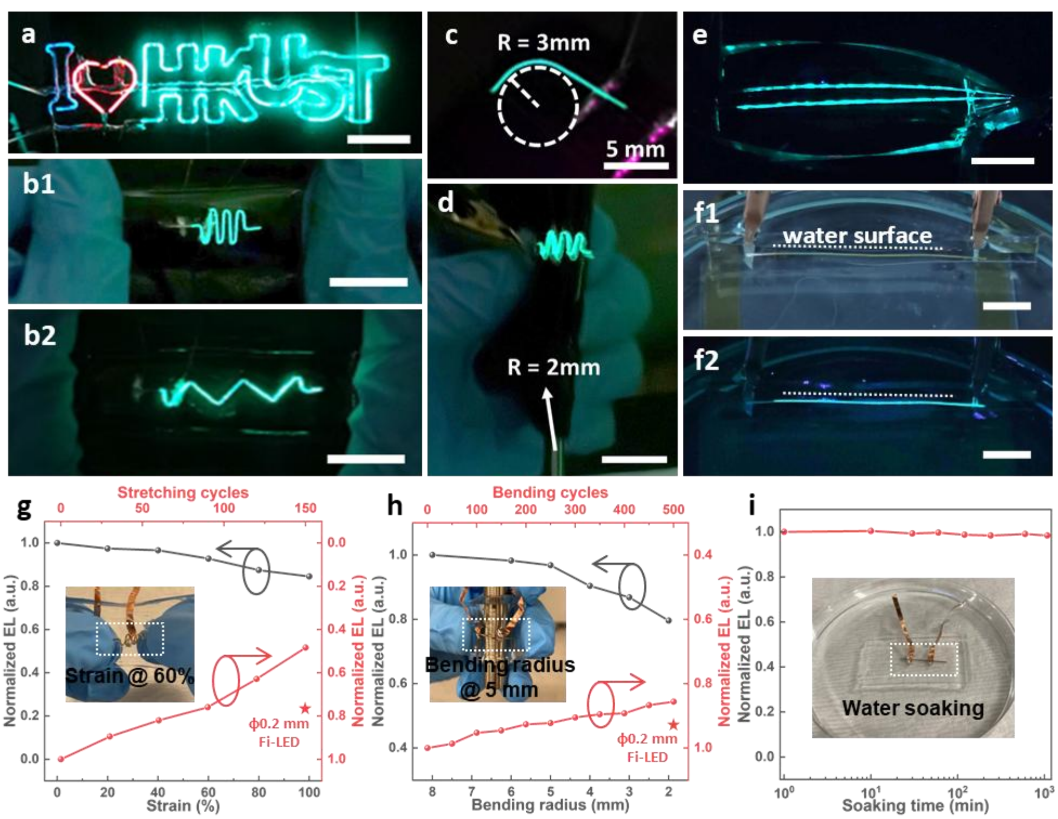

Figure 4. (a-f) EL characterizations in diverse scenarios: "I Love HKUST" pattern (a), stretchable device (b1, b2), bendable device (c, d), twistable device (e), and waterproof device (f1, f2). (g, h) Flexibility evaluations for PDMS-encapsulated Fi-LEDs: stretchability (g) and bendability (h) characterizations. (i) Aging test of the encapsulated device under continuous water soaking. All the unlabeled scale bars are 1 cm.

stretching cycles (star symbol in Figure 4g) and over 90% of its EL intensity following 500 bending cycles (star symbol in Figure 4h). Figure 4i presents the results of an aging test on the encapsulated device under water soaking. Benefiting from the superior PDMS encapsulation, the EL intensity of the Fi-LED remains unchanged even after continuous water immersion for over 1000 hours. These remarkable properties discussed above position the PeQW-based Fi-LED device as one of the highly appealing candidates for flexible and wearable lighting and display applications, with its potential integration onto fabrics further enhancing its appeal in this regard.

3. Conclusion

Overall, a fiber-shaped PAM template is utilized to grow PeQWs uniformly on Al fibers, forming structures spanning 1D, 2D, and 3D architectures. The unique two-step evaporation method enables Fi-LED device fabrication on a variety of unconventional frameworks. Ascribed to the malleability and plasticity of Al fiber, coupled with PDMS encapsulation for protection, the PeQW-based Fi-LEDs are successfully embedded into fabric, creating devices that are bendable, stretchable, twistable, and waterproof, which are essential for wearable applications. The facile and unique bonding and fabrication process here allows us to construct arbitrary architectures with multiple colors, aiming at future advanced lighting and display applications.

4. Acknowledgements

We acknowledge the support from the Material Characterization and Preparation Facility (MCPF) and the State Key Laboratory of Advanced Displays and Optoelectronics Technologies at HKUST. We acknowledge the assistance of Southern University of Science and Technology Core Research Facilities. Z. F. acknowledges the support from the New Cornerstone Science Foundation through the XPLORER PRIZE and Hong Kong Alliance of Technology and Innovation through BOCHK Science and Technology Innovation Prize. Funding: This work was supported by Hong Kong Research Grant Council (N_HKUST657/22, 16205321, 16309018, and 16214619), Shen Zhen Science and Technology Innovation Commission (JCYJ20170818114107730 and JCYJ20180306174923335), Innovation Technology Fund (GHP/014/19SZ), Guangdong-Hong Kong-Macao Intelligent Micro-Nano Optoelectronic Technology Joint Laboratory (2020B1212030010), and Foshan Innovative and Entrepreneurial Research Team Program (2018IT100031).

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