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**Air Stable Organic-Inorganic Perovskite Nanocrystals@Polymer
Nanofibers and Waveguide Lasing**

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ABSTRACT

Organic-inorganic hybrid perovskites have been considered as promising gain materials for lasing. Despite previous reports of lasing from nanocrystals, thin films and single crystals, the stability of perovskite lasers has been a challenge for its practical applications. Herein, we demonstrate a scalable strategy to prepare ultrastable perovskite@polymer hybrid fibers by employing a facile emulsion electrospinning approach. During the electrospinning process, polymethyl methacrylate (PMMA) first solidifies into an outer shell layer. Meanwhile, emulsion drops containing poly(vinylidene fluoride) (PVDF) and perovskite precursor are pushed inward and evolve into perovskite nanocrystals covered by PVDF. The PMMA with smooth surface benefits the light transport and the water-resistant PVDF blocks the moisture. The MAPbBr₃ perovskite-embedded fibers can emit intensive light after storage in humid ambient environment (relative humidity >60%) or even in water. We demonstrate amplified spontaneous emissions from the fibers network and waveguide lasing from chopped single fiber.

Keywords: Perovskite nanocrystals; perovskites polymer nanofibers; air stability; emulsion electrospinning; waveguide lasing

Hybrid organic-inorganic perovskites have attracted wide attentions owing to their potent optoelectronic applications associated with their unique features of excitons such as long carrier lifetime and diffusion length.^{1, 2} Since the discovery of perovskite as a promising gain material for lasing, many approaches have been developed to realize lasing from planar cavities, such as faceted single crystals and distributed feedback resonators.³⁻⁵ Despite the demonstration of perovskite lasers, the stability is still a big challenge.⁴ Particularly for nanoscale lasers for optical information processing and communication, the devices have to be handled under an inert atmosphere to avoid decomposition of the hybrid perovskite. This hinders the applications of perovskite lasers for commercialization.^{6, 7}

Polymer nanofibers with diameters of tens to hundreds of nanometers are versatile

platforms for light manipulation on the nanoscale.⁸⁻¹¹ Polymer waveguides in the form of optical nanofibers are considered as promising building blocks for photonic circuitry and integration.^{12, 13} Addition of gain materials (e.g., quantum dots or dye molecules) to polymer nanofibers allows one to achieve organic lasers with additional desirable features such as flexibility and low fabrication cost.^{14, 15} Inspired by this, we intend to deploy organic-inorganic hybrid perovskite as the gain material within the polymer cavity to achieve air-stable nanoscale laser. In this configuration, the polymer functions as both the protector and optical cavity. For this purpose, it requires that the polymer be water tolerant and the perovskite gain be uniformly dispersed inside the polymer.

Among various methods for preparing polymer fibers, electrospinning, based on the stretching of polymer solution by a strong electrostatic field, is a low cost and high throughput technology. Recently, electrospinning method has been applied for perovskite materials. Mathur and co-workers fabricated $\text{MeNH}_3\text{PbI}_3$ nanofiber mats under inert conditions.¹⁶ In another work, Tsai et al exploited the core-shell electrospinning approach to fabricate perovskite@polymer core-shell nanofibers, which are highly stable in air and water environments.¹⁷ However, no amplified emission or lasing has been realized from these perovskite-containing nanofibers.

To effectively couple the organic-inorganic perovskite gain materials with electrospun polymer fibers, it is a prerequisite that the polymer is waterproof and can prevent moisture attack. Unfortunately, when a single polymer is used for electrospinning, water in air can penetrate into the polymer due to the evaporative cooling effect which leads to the formation of pores in the polymer and adsorption of water.^{8, 18, 19} **The pore formation is more obvious when the electrospinning is conducted in a high-humidity atmosphere.** In addition, when one type of polymer is used, the perovskite will be dispersed within the entire fiber, and those distributed near the fiber surface is sensitive to moisture, leading to deterioration of optical property. For example, in our experiments, we have separately electrospun poly(vinylidene fluoride) (PVDF), polymethyl methacrylate (PMMA) and polyvinyl pyrrolidone (PVP) with methylammonium lead bromide (MAPbBr_3) perovskite precursor. But none of these composite fibers show intensive and stable luminescence.

Herein, we develop a two-polymer derived emulsion electrospinning process (**Figure 1a**),

which is able to produce ultrastable perovskite@polymer nanofibers with lasing action (denoted as L-PPNFs). Based on control experiments, we propose that the formation process of core-shell L-PPNFs involve two steps: (i) the solvent on the fiber surface evaporates rapidly during fiber stretching. PMMA tends to solidify first and forms an outer protective layer. In the meantime, the rapid solvent evaporation contributes to the phase separation of the incompatible PMMA/PVDF jet, leading to the formation of a core-shell structure in which PMMA covers PVDF. (ii) The emulsion drops containing perovskite precursor and PVDF are pushed inward during solvent evaporation and stretched to individual particles by the surface forces from PMMA (Figure 1b). Eventually, the polymer nanofibers with perovskite nanocrystals embedded near the center are obtained (Figure 1c). The resultant L-PPNFs are very stable and remain in water for one month without any evident decomposition. It should be stressed that all the processes from preparation to measurement are conducted in air with a high humidity (more than 60%).

Figure 1. Electrospraying synthesis of MAPbBr₃ perovskite@polymer nanofibers (L-PPNF). a) Schematic of the emulsion electrospraying; b) Emulsion droplets are pushed inward during solidification of the polymer fiber. Perovskite nanocrystals are formed after full an evaporation of solvent; c) Schematic of the final L-PPNF nanofibers.

For photonic applications, the waveguide loss is one of main obstacles constraining their optical performance. Among various types of polymer nanofibers, PMMA is proven to be ideal waveguide material with less than 0.1 dB mm⁻¹ loss.^{10, 20, 21} PVDF with affluent fluorine has excellent waterproof performance due to its hydrophobic nature.²² Therefore, we employed PMMA as the matrix and PVDF solution containing perovskite precursor to form

emulsion droplets. Both pristine polymer solutions appear clear. After they are mixed for a while, a cloudy emulsion solution is obtained, which is due to immiscibility and large difference in solution property between PMMA and PVDF in the DMF solvent (**Figure S1**, Supplementary Information). The microscopic images in **Figure S2** (Supplementary Information) also clearly show that the emulsion droplets (about 10 μm) are formed by mixing PMMA solution to perovskite precursor@PVDF solution. The volume ratio of PMMA to perovskite precursor@PVDF solution is 2:1, and the emulsion droplets are composited by perovskite precursor@PVDF solution.²³ After the solvent is evaporated, we obtained perovskite particles covered by PVDF (**Figure 2a and b**).

Figure 2. Morphology of the perovskite@polymer nanofibers. a) and b) TEM images of L-PPNFs. c) TEM image of nanofibers obtained using PMMA, for which no obvious perovskite nanocrystals are formed. d) TEM image of perovskite@PVDF nanofibers. There are plenteous irregular-shaped perovskites inlayed in the PVDF nanofibers which also have a coarse surface. e) Photographs of the L-PPNF after immersion in water for different times under UV lamp illumination. f) Photographs of L-PPNF collected on a piece of A4 paper and the corresponding fluorescence image under UV lamp illumination, showing the sample uniformity. g) SEM image of L-PPNF on a large scale.

For a typical composite fiber, the MAPbBr_3 perovskite particles are distributed in the core of nanofibers with a diameter around 170 nm and the thickness of protective layer is about 65 nm (**Figure S3**, Supplementary Information). Selective area electron diffraction (SAED)

patterns for L-PPNF exhibits diffused rings that are typical of random-oriented nanocrystals (**Figure S4**, Supplementary Information). It is interesting that the perovskite nanoparticles are mainly concentrated in the core area of the fibers rather than being randomly distributed in the whole fiber body. To explain this, we propose that the solidification of matrix should start from the outer layer and then continuously push the emulsion droplets inward (**Figure 1b**).²⁴ In this manner, the perovskite nanoparticles are fixed in the core after the total evaporation of solvent (**Figure 1c**). AFM topography measurements further confirm the fiber morphology and surface roughness of L-PPNF. **Figure S5** (Supplementary Information) clearly reveals the cylindrical profile of the L-PPNF with perfect circular section (The height equals the FWHM, thus indicating the diameter of the cylinder). The surface roughness as averaged along the L-PPNF length, is less than 1.5 nm.

To verify the importance of the emulsion, both perovskite@PMMA and perovskite@PVDF composite nanofibers were also fabricated using similar electrospinning technique (without emulsion). In the case of PMMA, the surface of the nanofibers is smooth (**Figure 2c and S6**), which is beneficial to a waveguide cavity.²⁵ However, it is difficult to detect any nanoparticles from the TEM images, indicating that the crystallization of perovskite was suppressed during the PMMA solidification process. The content of gain material is too low in the PMMA nanofibers to observe strong luminescence (see discussion below). In the case of perovskite@PVDF nanofibers, plenty of nanoparticles can be found randomly distributed inside the whole fibers (**Figure 2d and S7**). In addition, the content of perovskite nanocrystals in PVDF is obviously higher than that in PMMA due to the different chemical interactions, where the hydrogen bonds and lone pairs electrons of these polymers yield different degrees of coordination with either MA⁺ or Pb²⁺. However, the surface of PVDF nanofibers is rough. The coarse surface has been proven to be detrimental to lasing due to the relatively higher light scattering and waveguide loss.^{26, 27} Thus, the above control experiments prove that by employing emulsion electrospinning, the merits of two polymers can be combined to achieve a synergistic effect on waveguide cavity.

The obtained L-PPNF is very stable in air and water. A piece of free-standing L-PPNF membrane peeled off from the substrate (**Figure S8**) can emit considerable green light under UV illumination even after being submerged in water for one month, giving the credit to the

protection by the waterproofing PVDF (**Figure 2e**). The fluorescence emission is uniform across a standard A4 sized free-standing L-PPNF film (**Figure 2f**), in which the composite fibers have an average diameter of ~420 nm (**Figure S9**, Supplementary Information) and length up to millimeters (**Figure 2g**).

X-ray characterizations were conducted to study the chemical structures of the samples. In the X-ray diffraction (XRD) pattern of perovskite@PMMA, the peaks of perovskite are undetectable, while very weak peaks of perovskite can be observed in the case of perovskite@PVDF. But in L-PPNF, several peaks can be observed between 30 and 50° that can be indexed to the (210), (211) and (220) planes of the MAPbBr₃ (**Figure S10**). This confirms that the emulsion electrospinning favors the formation of perovskite nanocrystals. The X-ray photoelectron spectroscopy (XPS) was also performed to confirm if there are perovskite nanocrystals on the surface of the fibers and possible atomic interactions (**Figure S11**, Supplementary Information). Four elements (Pb, C, O and F) were probed, where the signal of Pb is very weak and the N and Br signals are nearly invisible. This is in accordance with the earlier observation that the perovskite nanocrystals are located below the surface and also beyond the detection limits of XPS (TEM image shows that the thickness of protecting layer is about 65 nm, while the probing depth of XPS is only around 10 nm). Trace Pb on the surface can be related to the strong interactions of Pb with O and F in the polymer chains.

It is expected that the type of polymer should affect the optical properties of the composite nanofibers. As mentioned above, photoluminescence emission is undetectable from perovskite@PMMA nanofiber because of the absence of waterproof PVDF. Although evident photoluminescence is observed for the perovskite@PVDF nanofibers, the intensity is very weak and the fluorescence image displays as a necklace-like appearance, which is thought to be caused by light scattering by the rough surface. In addition to scattering, intermittent emission (i.e., blinking) also occurs in the perovskite@PVDF nanofibers, that is likely related to trapped charges induced auger recombination process.^{28, 29} The blinking will largely reduce the possibility of stimulated emission and also disturb the continuous coupling with the polymer wire resonator.³⁰

Figure 3. Optical characterization of MAPbBr₃ L-PPNF. a) Photoluminescence spectra of four different samples: perovskite@PVDF nanofiber with blinking (red curve), and L-PPNF with different fiber widths (green curve: ~200 nm; black curve: ~2 μ m; blue curve: L-PPNF ~450 nm). Excitation wavelength: 473 nm; power: 10 μ W; acquisition time: 10 s. Fluorescence images of three corresponding samples are presented in background. b) Comparison of room-temperature photoluminescence, absorption, and lasing spectra of L-PPNF. Photoluminescence peak intensity decay for c) fresh L-PPNF and d) L-PPNF after storage in the air ambient for 10 days.

Compared to the control samples with single polymer, the co-blending polymer nanofibers obtained by emulsion electrospinning exhibits significantly enhanced and more homogeneous luminescence emission, which can be seen from both photoluminescence spectra and the fluorescence images (**Figure 3a and S12**, Supplementary Information). The emission intensity is found to be dependent on the perovskite concentration and polymer fiber diameter. For the optimized sample (L-PPNF in **Figure 3a**), the photoluminescence intensity is nearly 20 times that of the perovskite@PVDF nanofibers (blinking sample). In the optimized case,

the core area forms higher quality and relatively larger nanocrystals (average diameter around 11 nm; **Figure S3**, Supporting Information), where the confined excitons can easily emit light via radiative combination.³¹ Additionally, the polymer shell acts as both the optical cavity and protector. The AFM topography (**Figure S8**, Supplementary Information) proves that the polymer fibers have high surface smoothness (roughness <1.5 nm), which is favourable for the optical cavity function.³² The absorption spectrum (**Figure 3b**, the orange track) taken from high density L-PPNF networks on a SiO₂/Si substrate shows clear excitonic peak on the absorption edge, indicating high optical quality of these wires. The photoluminescence spectrum is slightly red shifted due to stokes shift, which also implies high efficiency of absorption.³³ We also studied the photostability of the L-PPNF in ambient air condition by continuously illuminating the samples with 350 nm excitation. As shown in **Figure 3c**, L-PPNF could keep about 96% of the initial luminescence intensity upon the continuous exposure to 350 nm excitation for 2 h. After 10 days storage in lab atmosphere (humidity around 80%), the L-PPNF retained 93% of the initial intensity under the same illumination condition (**Figure 3d**). Such a high air stability assures its application in photonic devices.

Based on the high absorption efficiency and optical quality, the population inversion of the L-PPNF was also studied by power-dependent photoluminescence measurement with a pulsed laser (400 nm, repetition rate 80 MHz) excitation focusing on a local area of the MAPbBr₃ nanocrystals embedded in the polymer fibers. As shown in **Figure 4a**, obvious amplified spontaneous emission (ASE) can be observed when the pump fluence increased to about 20 $\mu\text{J cm}^{-2}$. The inset of **Figure 4b** shows the transition from spontaneous emission to ASE. The first three points showed very weak emissions because the pump fluence was still below the trapping densities.³¹ To realize possible random lasing, the excitation was then focused onto an enlarged area (by defocusing the microscope) with a high-density fibers network to ensure that a virtual cavity is pumped as a whole. For this purpose, a pulsed laser of 425 nm (repetition rate 4 kHz) was used for excitation. Random lasing was observed only when the pump fluence was extremely high as $\sim 260 \mu\text{J cm}^{-2}$ where the spontaneous emission background is still visible (**Figure S13**, Supplementary Information). Obviously, the loss is high in the case of random cavity where the stimulated emissions will experience abundant inter-fiber interface.

Figure 4. Amplified spontaneous emission (ASE). a) Power-dependent photoluminescence spectra of the L-PPNF bundles, demonstrating the occurrence of ASE. b) Plots of light emission intensity in response to pump fluence. The crossing point in the inset figure refers to the onset of population inversion. Random lasing can be observed at extremely high pump fluence of $430 \mu\text{J}/\text{cm}^2$ (Figure S12, Supplementary Information).

To achieve better cavity from individual wires, we designed a direct dry cut method to peel out L-PPNF fragments. In this process, a tungsten probe with ultra-sharp tip ($\sim 5 \mu\text{m}$ size) was used to scratch on the L-PPNF networks and fetch isolated nanofibers on a glass substrate. The isolated fiber with a length around $20 \mu\text{m}$ was studied as a waveguide laser.³⁴

Figure 5a visually presents the lasing image when the pump fluence increases across the threshold. From the images, one can observe spotty emissions from both ends of the fiber with significantly suppressed spontaneous emission background. The photoluminescence spectra of a typical L-PPNF (length $\sim 20 \mu\text{m}$ and diameter $\sim 400 \text{ nm}$) was also measured at a pumping fluence ranging from 6.49 to $68.2 \mu\text{J cm}^{-2}$ (**Figure 5b**), illustrating the occurrence of sharp and intensive emissions. To see the lasing effect more clearly, the integrated output emission and the full width at half-maximum (FWHM) over the whole spectra range was plotted as a function of pumping fluence (**Figure 5c**), which clearly indicates the evolution from spontaneous emission to lasing behaviour with a sharp kink at the threshold around $30.0 \mu\text{J cm}^{-2}$.

Figure 5. Waveguide lasing of single L-PPNF. a) Bright-field image (left one) and dark-field images (right three, with a 532 nm filter) of an isolated fiber at increasing pump fluences. Scale bar: 5 μm . b) Power-dependent emission spectra of the isolated fiber show the appearance of lasing. c) Plot of emission intensity of the peak at 538.3 nm and FWHM as a function of pump fluence (peak fitting is provided in Figure S13, Supplementary Information). d) TRPL decay kinetics under pump fluence below ($P \sim 0.79 P_{\text{Th}}$, blue) and above ($P \sim 1.25 P_{\text{Th}}$, red) the threshold. The lifetime is about 2.9 ns for the spontaneous emission below P_{Th} and ≤ 70 ps for the lasing process above P_{Th} . The black curve is the TRPL with a lifetime ~ 15 ns at very low pump fluence ($P \sim 0.1 P_{\text{Th}}$).

From the spectra measured above the lasing threshold, the mode distance ($\Delta\lambda$) of lasing peaks is around 2.1 nm, which is higher than the estimated value (~ 1.2 nm) from the equation $\Delta\lambda = (1/L)[(\lambda^2/2)(n - \lambda dn/d\lambda)^{-1}]$ for a Fabry–Pérot cavity, where L is length of the cavity (ca. 20 μm in this case), n is refractive index of the polymer (ca. 1.47 as average between PMMA and PVDF). The large deviation could be related to scattering loss along the polymer fiber body and the nonuniform distribution of the gain material within the cavity. The cavity mode of 538.3 nm has a FWHM of ~ 0.69 nm, corresponding to a quality factor (Q-factor) of ~ 780 (see details in **Figure S14**, Supplementary Information). However, when the pump fluence reaches to a high value of 58.8 $\mu\text{J cm}^{-2}$, there appear two additional modes

at 544.3 and 546.5 nm (Figure 5b). A possible reason is that the low refraction index of the polymer fiber surface allows certain leakage of the multi-modes at high pump fluence. The lasing behaviour was further studied using time-resolved photoluminescence (TRPL) measurements (Figure 5d). The exciton lifetime (τ_{SPE}) extracted from fitting is 15 ns at very low excitation densities ($P=0.1P_{th}$, black dots and fit line). At the higher excitation density ($P=0.8P_{th}$, blue dots and fit line), τ_{SPE} decreases to 2.9 ns, where the stimulated emission starts to compete with and suppress the spontaneous emission. As the pump fluence further increases beyond the threshold, an ultra-short emission lifetime ($\tau_{Lasing}\sim 67$ ps) is obtained, corresponding to the lasing emission. Since the stimulated emission time constant is much less than τ_{SPE} , most of the energy is efficiently dissipated to lasing.

In conclusion, we have demonstrated successful synthesis of air-stable perovskite nanocrystals embedded within polymers using emulsion electrospinning and the realization of waveguide lasing from MAPbBr₃ perovskite@polymer composite fibers. The L-PPNFs can survive in water for one month. Therefore, all the processes, from fabrication to measurement, can be conducted in normal lab atmosphere with high humidity. To our knowledge, this work is first to realize air-stable perovskite nanolaser by a facile and low-cost electrospinning method. Comparing to perovskite single-crystal lasers, L-PPNFs are flexible and have more promising application potential in flexible photonics devices. This strategy may open the door to the synthesis of a broad class of stable photonic systems based on hybrid perovskites.

Experimental

Fabrication of perovskite@polymer nanofibers. Solution A for synthesizing perovskite@PMMA fibers: 0.6 g PMMA ($M_w\sim 120,000$ by GPC) was added into 5 ml DMF solvent under stirring at 60 °C. Solution B for synthesizing perovskite@PVDF fibers: 0.05 mmol perovskite precursor and 0.5 g PVDF ($M_w\sim 534,000$ by GPC) were dissolved into 5 ml DMF. Solution C for L-PPNFs: 1 ml solution B was added into 2 ml Solution A without perovskite and then the emulsion solution was obtained after vigorously stirring. For electrospinning, the high voltage of 12 kV was applied between the needle and collecting substrate. The working distance was ac. 10 cm and the solution feeding rate was 1 ml/h.

Characterizations. Morphology and size of samples were examined by SEM (Hitachi SU-70) and TEM (JEOL 2100 FEG). The elemental analysis was carried out by multifunctional X-ray photoelectron spectroscope (Shimadzu, AXIS ULTRA DLD). X-ray diffraction patterns were obtained using an X-ray diffractometer (XRD Shimadzu Thin Film). AFM experiments were performed in tapping mode under ambient conditions (Dimension ICON SPM system, Bruker, USA).

ASE and lasing measurement. A 400 nm pulse excitation was used to study ASE of the polymer coated perovskite. The 400 nm pulse was generated by frequency doubling of the 800 nm output from a mode-locked Ti/Sapphire oscillator (Spectra-Physics, 80 MHz repetition rate, 100 fs pulse width). Measurements were performed using a micro-Raman spectrometer (Horiba Jobin–Yvon HR800) in reflection geometry, and pump laser was introduced by a 100× microscope. The backscattered emission was collected by the same objective, and then analyzed by a liquid nitrogen-cooled CCD detector. Lasing measurement was conducted using the same setup, except for the 425 nm pulse excitation generated from an optical parametric amplifier (Spectra-Physics TOPAS, 1 kHz, 100 fs). A 20× microscope was used to introduce excitation and collect lasing signal. The excitation laser was focused to a spot size of ~ 30 μm in diameter to assure sufficient energy injection from the whole nanofiber.

Time-resolved photoluminescence spectroscopy (TRPL). The fluorescence lifetime measurement was performed using a confocal microscopy with 20× microscope. 400 nm pulse excitation (800 kHz, 100 fs) was introduced to the sample, and the emission was analyzed by a single photon avalanche diode (SPAD) to resolve the time decay. 458-nm long-pass and 720-nm short-pass filters were used to select the photoluminescence emissions.

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TOC Entry

Hybrid perovskite nanocrystals embedded within polymer nanofibers have been fabricated via a facile emulsion electrospinning approach, which are stable in 80%-humidity air for up to a month. Stable luminescence and waveguide lasing have been achieved from MAPbBr₃ nanocrystals@polymer nanofibers.