Continuous-wave operation in directly patterned perovskite distributed feedback light source at room temperature

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We report a directly patterned perovskite distributed feedback (DFB) resonator and show narrow amplified spontaneous emission (ASE) at pump powers as low as 0.1 W/cm² under continuous-wave (CW) optical pumping conditions at room temperature. Compared to the pristine thin film photoluminescence spectrum, a 16-fold reduction in emission linewidth in the MAPbI₃ DFB cavity was observed. The direct nanostructuring of perovskites was achieved by thermal nanoimprint lithography. Our findings pave the way toward realizing CW pumped perovskite lasers at room temperature and energy-efficient perovskite light sources. © 2018 Optical Society of America

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A crucial component in high-performance photonic integrated circuits is a miniature light source that is efficient, silicon-compatible, electronically addressable, and economical. To this end, solution-processed hybrid perovskites have emerged as the material candidate that may satisfy all of the above requirements [1–3]. Additionally, they have strong optical absorption [4], long carrier diffusion length [5], high carrier mobility [6], broad wavelength tunability covering the entire visible spectrum and into the near-infrared (near-IR) spectrum, and support cost-effective fabrication [7]. However, a major roadblock in the progress of perovskite light sources is the inability to directly pattern them. Because of material instability under moisture and solubility in many solvents, perovskite is incompatible with optical or e-beam lithography techniques due to the use of solvents during the resist development process [8].

In existing literature, perovskite lasers based on periodic structures (the photonic crystal type) are typically constructed using a spin-coating or evaporation process on pre-patterned structures [9–14]. On the other hand, perovskite lasers based on isolated cavities (such as microdisk, micropalette, and nanowire) are constructed using self-assembly or solution-phase growth methods [15–23]. Other laser designs, such as the vertical cavity surface emitting laser (VCSEL) with thin film perovskite [24,25] or random laser with scattering films [26], are also explored. However, the above-mentioned approaches all lack dimension control, repeatability, uniformity, and scalability. Patterning of perovskite by focused ion beam [27] has been attempted, but impurities are inevitably introduced into perovskites during the milling process.

Nonetheless, with cavities formed by these methods, amplified spontaneous emission (ASE) and lasing have been observed in optically pumped devices, all with ultrashort pulses (femtosecond or picosecond). In [9], the authors demonstrated a nanosecond pulse pumped MAPbI₃ perovskite laser at cryogenic temperatures. It was speculated that the pump power density required for MAPbI₃ perovskites to lase under continuous-wave (CW) pumping would be ~14 KW/cm² [24]. Recently, an ASE in a CW optically pumped MAPbI₃ vertical cavity emitter was demonstrated, with a nine-fold linewidth reduction compared to the material photoluminescence (PL) spectra, marking a major step toward realizing a CW optically pumped perovskite laser [25].

One laser design that promises single-mode emission at a user-defined wavelength is the distributed feedback (DFB) cavity. Thus far, there have been several demonstrations of a perovskite DFB laser [10,13,14]. Although a common approach for fabricating organic DFB lasers is the direct nanoimprinting of the soft gain medium [28], because perovskites are hard, ionic-based materials without a glass transition behavior, nanoimprint cannot be readily applied to perovskite. Instead, these DFB lasers were created by depositing the perovskite film on top of the patterned substrate.

In this Letter, we report—for the first time to our knowledge—a directly patterned MAPbI₃ DFB resonator, and show narrow ASE under CW pumping at room temperature.
At pump powers as low as 0.1 W/cm², a 16-fold linewidth reduction compared to a material PL is achieved.

The nanostructuring is achieved with our recently demonstrated thermal nanoimprint lithography (NIL) of perovskites [29]. Figure 1(a) sketches the principle of NIL to pattern perovskites. Figure 1(b) shows SEM comparisons of a spin-coated MAPbI₃ thin film (top) and the MAPbI₃ structure after NIL (bottom). While polycrystalline MAPbI₃ with a grain size of a few hundred nanometers is obtained in the pristine thin film, the imprinted structure shows a much larger grain size and better uniformity. In fact, the smoother surface and larger grain size is a consequence of the material morphology improvement during NIL [29,30], which is helpful in reducing scattering loss in laser structures. Figures 1(c) and 1(d) show the perspective view of the SEM and AFM of an imprinted perovskite DFB cavity, respectively. The clearly defined gratings and smooth surface can be observed. The depth of the grating groove is measured to be 161 nm by AFM.

To create the perovskite DFB cavity, we first fabricated an SiO₂ mold for NIL by e-beam lithography with HSQ resist. We created a Cr hard mask for the subsequent etching of the SiO₂ because Cr has excellent selectivity for SiO₂ etching. To obtain a straight and smooth mold sidewall, SiO₂ was etched using inductivity coupled plasma (ICP) etching, followed by a Cr strip-off to form the mold for NIL. Lastly, the mold was coated with an anti-adhesion monolayer of perfluorodecyltrichlorosilane (FDTS) to prevent perovskite from sticking to the mold after NIL. Next, MAPbI₃ thin film was spin-coated onto an Si substrate with 1 μm thermal SiO₂, then the perovskite DFB structure was subsequently formed by thermal NIL. The details of the MAPbI₃ thin-film preparation and NIL process can be found in our earlier work [29]. The resulting device has an area of 100 μm by 100 μm.

The device was optically pumped with a 355 nm Nd:YVO₄ laser under CW operation, with an output power in the range of a few to two hundred μW. The pump beam was projected onto the sample through a UV microscope objective with a numerical aperture of 0.13 resulting in a pump spot size of 100 μm. The objective lens was also used to collect the emitted light in reflection mode. Using a cascaded 4-f imaging system in conjunction with a pump filter, the collected light was then directed into a spectograph with a cooled Si detector. Figure 2(a) shows the room temperature PL spectra of the device [with SEM in Fig. 1(c)] measured at various CW pumping levels. For all pump powers, we observed a broad higher-intensity peak close to the center of MAPbI₃ PL spectrum, as well as a narrow lower-intensity peak, which is at the edge of the MAPbI₃ PL. The full width half-maximum (FWHM) of the ASE peak at 813.6 nm is 2.4 nm. Interestingly, the narrow FWHM remains constant for all different pump powers employed, as shown in the inset of Fig. 2(a). Note that the power densities employed here are a few orders of magnitude lower than the reported ASE and
lasing thresholds [24,31]. When the pump power density is reduced below 0.1 W/cm², the emitted light is below the noise level of the detector and, therefore, cannot be detected. Thus, it is our speculation that the ASE threshold is lower than 0.1 W/cm². Thanks to the high-quality (Q) factor of the cavity resonance at ∼813 nm, the FWHM of the ASE peak collapsed to 2.4 nm from 38 nm of PL spectrum, as shown in Fig. 2(b).

Although a low ASE threshold was speculated, no lasing behavior was observed when the pump power was increased further. We attribute the absence of lasing at high powers to two factors. First, the cavity resonance at 813.6 nm is at the edge of the MAPbI₃ PL spectrum [Fig. 2(b)], resulting in a minimal amount of material gain. Second, self-heating and thermal instability are significant under CW pumping at room temperature. With the poor thermal conductivity of <0.5 W/mK in perovskites [32], large Auger recombination, decreased material gain coefficient, and material degradation are to be expected. Indeed, we observed a slight blue shift of ∼1 nm in the emission spectrum as pump power was increased, in agreement with the reported trend in the literature [33]. It is worth noting that there is another cavity resonance at ∼780 nm, evidenced by the broad ASE peak atop the PL spectrum. However, this mode has a much lower Q factor than that at ∼813 nm.

Figure 3 shows the simulated DFB resonator behavior. A dispersive refractive index profile from that of [34] is used, but scaled to account for other reported MAPbI₃ values [11,35] resulting in a refractive index of n ≈ 2.62 at 800 nm. Note that an infinite number of periods, zero scattering loss, and smooth surface were assumed in simulation and, as a result, we expect the actual Q factor of the device to be much lower than simulated. Figure 3(a) depicts the side view of the structure. For the DFB dimension in Fig. 1(c), namely, an air trench width w₁ = 70 nm, grating period a = 360 nm, grating height b₁ = 160 nm, perovskite residue thickness b₂ = 70 nm, Fig. 3(b) shows the transmission spectrum of the four highest-Q modes at near-normal incidence (incident angle 1°). A high-QTE₁ mode and a moderate-QTE₂ mode reside at the edge and close to the center of the PL spectrum, respectively. The resonance wavelengths of the TE modes are in close agreement with the experimentally observed ASE peaks in Fig. 2(b). No ASE peak from TM modes was observed experimentally, because they are either too low in Q factor or too close to the PL edge. For a given perovskite composition (in this case, MAPbI₃), the optimization of the emission wavelength and cavity Q factor can be accomplished by tuning the air trench width (w₁), grating height (b₁), and period (a), as well as the perovskite imprint residue layer thickness (b₂). For the highest-QTE₁ mode, Figure 3(c)–3(d) shows the Q factor and resonant wavelength’s dependence on the grating period and air trench width, with all other design parameters kept the same as in Fig. 3(b). The increase of the ASE wavelength with an increasing grating period was also observed experimentally, as shown in Fig. 4.

We note that while cavity design can indeed be optimized by changing NIL mold dimensions, such as air trench width...
and grating period, each iteration requires the fabrication of a new mold. Another degree of freedom the DFB laser design via NIL provides is that one can tune the cavity behavior by simply adjusting the perovskite residue thickness (62), without any additional nanofabrication steps. For an air trench width of 70 nm and 105 nm, Fig. 4 also describes the dependence of the TE<sub>1</sub> mode ASE wavelength as a function of the perovskite residue thickness.

In summary, we demonstrated the first, to our knowledge, directly patterned perovskite DFB resonator and showed room temperature operation of the device under CW optical pumping. A narrow ASE linewidth of 2.4 nm—a 16-fold reduction from MAPbI<sub>3</sub> PL linewidth—was achieved with pump powers as low as 0.1 W/cm<sup>2</sup>, indicating the superior cavity behavior. Although self-heating, thermal instability, misalignment of cavity resonance, and perovskite gain spectrum prevented lasing action in the device, our approach to directly pattern perovskite is a promising method for energy efficient perovskite lasers. With improved thermal stability [33], material stability [36], better alignment of the cavity mode with perovskite PL, and an optimized NIL process, low-threshold CW optically pumped and even electrically pumped lasing are possible. Additionally, using the cost-effective and massively scalable nanoimprint lithography, various perovskite optoelectronic devices can be realized, thus paving the way to all-perovskite, silicon-compatible photonic integrated circuits.

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