Lasing from Halide Perovskites

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***Abstract* — Organic-inorganic halide perovskites (*e.g.*, $CH_3NH_3PbI_3$) possesses exceptional optoelectronic properties for photovoltaics. Amazingly, this material system also has outstanding optical gain properties ideal for lasing applications. Two years had elapsed since the discovery of amplified spontaneous emission and lasing from these perovskite thin films and nanostructures. Herein, we briefly review its development, the state-of-the-art and the prospective outlook of this new lasing medium.**

Keywords* — *organic-inorganic halide perovskites, thin films, plateletets, nanocrystals, amplified spontaneous emission and lasing

I. Introduction

Solution-processed organic-inorganic halide perovskites (*e.g.*, $CH_3NH_3PbI_3$) have garnered immense research interests because of their impressive >20% photovoltaic conversion efficiencies. These remarkable efficiencies stem from their outstanding optoelectronic properties of long balanced electron-hole diffusion lengths [1] and low defect densities [2]. Surprisingly, these halide perovskite materials are also excellent optical gain media. Given their long range ambipolar charge transport properties, this raises an exciting possibility of realizing electrically-driven lasing with a solution processed gain media. The first report of ultralow threshold amplified spontaneous emission (ASE) from $CH_3NH_3PbI_3$ thin films and microcavity lasing from $CH_3NH_3PbI_3$ crystals [2] occurred amidst the fervent photovoltaic efficiency race in early 2014. Since then, optical gain have also been demonstrated in perovskite micro/nano-structures [3-5] and colloidal perovskite nanocrystals [6-8]. In view of this rapidly expanding field, we will highlight some of the key works on perovskite lasing.

II. Background and State-of-The-Art

A. Seminal Works

Room-temperature (RT) optically-pumped ASE and lasing was first demonstrated in 65 nm-thick thin films $CH_3NH_3PbX_3$ perovskites (X = Cl^-, Br^-, or I^-), with threshold fluence as low as 12 ± 2 μJ/cm^2 (or carrier density ~1.7 × 10^{18} cm^{-3}). This relatively low ASE threshold is attributed to their large absorption coefficients, small capture cross-sections of trap states, low bimolecular recombination and "slow" Auger recombination properties. Importantly, $CH_3NH_3PbI_3$ is an extremely durable gain medium whose ASE intensity remained invariant (*i.e.*, with only 0.2% standard deviation about $I_{average}$) under 26 hours of sustained irradiation (*i.e.*, ~10^8 laser shots) with a 1 kHz Ti-sapphire regenerative amplifier. By straightforward combination of the different precursors, the composition of these perovskite films could also be easily tuned to achieve broad ASE wavelength tunability from ~390 nm to 790 nm. Furthermore, Xing *et al.* [2] was also the first to realize RT lasing from a $CH_3NH_3PbI_3$ crystal several tens of micrometers in size found in solution processed drop-casted thin films. Around the same period, Deschler *et al.* [9] demonstrated RT optically pumped lasing from $CH_3NH_3PbI_{3-x}Cl_x$ in a micro-cavity constructed using a sandwiched perovskite film between a commercial Bragg reflector mirror and a thin gold layer (with a threshold of 0.2 μJ per pulse using 400 ps pump pulse). These two key papers sparked a global interest in the optical gain properties of perovskites.

B. ASE/Lasing from Perovskite Thin Films

Stranks *et al.* [10] demonstrated further reduction of ASE thresholds by ~2 orders by sandwiching $CH_3NH_3PbI_3$ between a cholesteric liquid crystal reflector/gold back-reflector cavity. Perovskite lasing was also controlled using in-plane structuring of perovskite thin films on 2D patterns and 2D photonic crystal resonators. Saliba *et al.* fabricated perovskite distributed feedback (DFB) lasers with very low thresholds of ~0.32−2.11 μJ/cm^2 (using 1-ns pump pulses) by evaporating perovskite film onto a nano-imprinted polymer resist, The lasing can be tuned between 770 - 793 nm by varying the grating periodicity. Chen *et. al.* [11] most recently demonstrated perovskite photonic crystal (PhC) devices with threshold densities of 68.5 ± 3.0 μJ/cm^2 and an approach to scaling 2D perovskite thin film lasers in a multi-element pixelated array of microlasers for possible projection display applications.

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C. *Lasing from Micro/nano-structures and Colloidal Perovskites*

RT lasing in perovskite micro-/nanostructures was first reported by Zhang *et al.* [3] in highly crystalline organic−inorganic perovskite $CH_3NH_3PbI_{3-x}X_x$ (X = I^-, Br^-, and Cl^-) *platelets* with nanoscale thickness (10−300 nm) and edge length of several to tens of micrometers. Q-factors as large as ~1300 could be achieved. Likewise, Liao *et al.*[4] also reported the realization of WGM lasers from $CH_3NH_3PbBr_3$ single crystal square microdisks (prepared using a self-assembly method) with lasing thresholds as low as ~3.6 μJ cm^{-2} using 120-fs pump excitation.

The first report on perovskite *nanowire* (NW) lasers was by Zhu *et. al.* [5], who demonstrated low threshold RT lasing of ~220 nJ cm^{-2} (*i.e.*, carrier density of 1.5×10^{16} cm^{-3}) and a very large high Q-factor ~3600. Due to their ultra-compact physical sizes, highly localized coherent output, and efficient waveguiding, NW lasers are regarded as promising building blocks for fully integrated nanoscale photonic and optoelectronic devices. Each NW can act as a waveguide along the axial direction, while the two end facets constitute a Fabry-Perot cavity for optical amplification. Since then, perovskite NW lasers have also been demonstrated in several works [12, 13], signifying the great potential of this fledging field.

Colloidal perovskite *nanocrystals* (NCs) are also another promising variant of perovskite gain media. Recently, lasing from all-inorganic colloidal NCs of cesium lead halide perovskites ($CsPbX_3$, X = Cl^-, Br^-, and I^-) was reported [6-8]. Compared to the traditional metal-chalcogenide NCs, these perovskite NCs deliver prominent optical gain signatures that combines the advantages of both quantum dots and halide perovskites: namely, excellent photochemical and oxidative stability even without shell coating (*e.g.*, CdS in traditional CdSe/CdS quantum dots), high PL quantum yield (up to ~90%), narrow emission linewidths of 70–100 meV, precise and continuous tunable emission wavelength peaks from 410-700 nm via compositional control (*i.e.*, mixed halide Cl/Br and Br/I systems) and effect of quantum confinement. The potential of this sub-class of perovskite nanostructures is immense.

III. Summary and Outlook

In summary, organic-inorganic halide perovskites are a new family of optical gain media. Despite the success of optically-pumped lasing from perovskites, electrically-pumped lasing is still elusive. Challenges include the high current densities (>5kA/cm^2) needed for electrical driven lasing and their heating effects. Possibly, a combination of careful materials design, growth and device engineering are needed to tackle these issues in the form of resonator designs (*e.g.*, usage of light emitting transistors architecture with a DFB or PhC resonator) and usage of high-quality single crystalline thin films or nanostructures (e.g., nanoplatelets, NWs, NCs, *etc.*). Nonetheless, the prospects of perovskite gain media will continue to shine in the foreseeable future.

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