Continuous-wave lasing in halide perovskites

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Continuous-wave (c.w.) lasing means that the laser is operated under continuous optical pumping, which is believed to be a key stepping stone on the path to an electrically pumped laser diode and to applications in high density integrated optoelectronic systems [1]. Achieving continuous-wave lasing was thought to be challenging due to the intrinsic thermal damage and Auger recombination loss, especially in organic semiconductor materials [2,3]. In the past two years, with high optical absorption coefficient, low density of defects and especially the suppressed Auger recombination rate, halide perovskites nanostructures are emerging as one of the most promising gain materials for the development of continuous-wave operated lasing, which paves the way for the ultimate goal of electrically pumped laser devices [4,5].

Recently, published in *Nature Photonics*, Prof. Chris Giebink and co-workers reported the demonstration of optically pumped continuous-wave lasing in methylammonium lead iodide (MAPbI₃) distributed feedback cavity at low temperature of 100 K [6]. In this work, the authors explore the gain properties by depositing MAPbI₃ film onto a period grating etched into an alumina layer on a high-thermal-conductivity sapphire substrate to avoid the thermal damage (Fig. 1a). In addition, another tricky designing of the continuous-wave laser is the dynamic phase-change process of the MAPbI₃ film. As shown in Fig. 1b, at a substrate temperature of 106 K, the amplified spontaneous emission (ASE) from the MAPbI₃ shifts from 750 nm (orthorhombic phase emission) to 785 nm (tetragonal phase emission) within several hundred nanoseconds due to the tetragonal phase inclusions formed inside the orthorhombic lattice matrix, presumably because of local excitation light heating. They demonstrated that the photogenerated tetragonal phase inclusions are supposed to function as charge carrier recombination sinks similar to the classical semiconductor quantum

wells [7], which reduce the Auger recombination rate and other unexpected non-radiative recombination channels, thus enhancing population inversion and providing optical gain. Notably, the authors also found that lasing from the tetragonal phase inclusions at 106 K could be sustained for more than 1 h (Fig. 1c), while lasing death occurs for pure tetragonal-phase MAPbI₃ at $T > 160$ K [8].

From the intrinsic fundamental physics point of view, further study is required to reveal the mechanism on why this mixed-phase avoids the lasing death phenomenon that occurs in pure tetragonal phase MAPbI₃ at $T > 160$ K. Anyway, in this study, the concept of phase transition and inclusions proposed by Giebink and co-workers provide a useful strategy in the pursuit of an electrically pumped perovskite laser diode, which further reveals that the effort put in diminishing the defect states and other non-radiative recombination channels is critical for achieving c.w. lasing.

Indeed, lasing in a nanoscale oscillator cavity may not require population inversion. With significant exciton-polariton effect in perovskite materials, it is possible to achieve polariton lasing in the perovskite structures especially in a highly confined nanowire cavity with enhanced oscillator strength *via* reduced mode volume. Very recently, Prof. Xiaoyang Zhu's group from Columbia University has reported c.w. polariton lasing in CsPbBr₃ nanowires with low pump intensity [3]. They claimed that both c.w. and pulsed lasing originated from polariton modes based on the cavity modes analysis. The authors also investigated the temperature dependent lasing modes using the Hamiltonian for exciton-photon coupling, which provided further evidences for the low threshold polariton lasing. Although the conversion of polariton lasing to photon lasing is difficult to detect due to degradation of these CsPbBr₃ nanowires at high pump intensity, the work demonstrates that lead halide perovskite nanowires show promising applications in low

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Figure 1 (a) Cross-sectional scanning electron micrograph (SEM) of the second-order DFB laser architecture consisting of a MAPbI₃ film deposited on an alumina grating on a sapphire substrate. (b) Streak camera image of emission from the MAPbI₃ film under 920-ns-long rectangular pump pulse at the substrate temperature of $T=106$ K. ASE from the orthorhombic phase of MAPbI₃ is observed at early times but evolves within ~ 200 ns (see blow-up in the inset) into tetragonal-phase ASE, which is sustained until the end of the pump pulse. (c) Continuous-wave laser emission intensity recorded from a single excitation spot over more than 1 h. Reproduced with permission from Ref. [6], Copyright 2018, Nature Publishing Group.

power consumption c.w. coherent light sources.

Lastly, these inspiring breakthroughs in c.w. lasing from perovskites undoubtedly represent important steps towards electrically pumped devices. For the prospective of room temperature c.w. lasing, the polariton lasing process in in-organic halide perovskite nanowires is more promising than relying on the phase-change process, e.g. Xiong *et al.* [9] have reported the experimental realization of room-temperature polariton lasing based on a CsPbCl₃ nanoplatelet embedded in a distributed Bragg reflectors (DBR) microcavity. However, according to Zhu's work, room temperature c.w. polariton lasing of the CsPbBr₃ nanowires is still facing challenge mainly due to the thermal and light degradation. To this end, the most important task is to improve the nanowire quality since samples with high stabilities would be possible to achieve the conversion of c.w. polariton lasing to photon lasing under high pump density at room temperature. On the other hand, highly confined cavity with enhanced room temperature photon-exciton coupling strength is also worth to develop, which would give rise to high density polaritons for lasing. Basically, the exploitation of the composition dependent exciton-photon coupling strength in CsPbX₃ (X=Cl, Br, I) nanowires is also indispensable to give theoretical guidance for room temperature c.w. laser constitution. It shall be expected that these nice studies will inspire efforts towards achieving the ultimate goal of

electrically pumped laser devices.

Received 3 April 2018; accepted 4 April 2018;

published online 3 May 2018

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