# Photoluminescence and Amplified Spontaneous Emission in Quasi-2D and 3D Perovskite: Influences of Excitonic Versus Free Carrier Emission

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Quasi-two-dimensional (2D) perovskites are promising optoelectronic materials for display and lighting technologies due to their excellent luminescent properties [1]. Light-emitting diodes (LEDs) based on quasi-2D emitters have demonstrated external quantum efficiencies over 20% [2]. Meanwhile, the recent demonstration of optically pumped continuous wave lasing at room temperature [3], giving quasi-2D perovskites front-runner status for realizing electrically driven lasers.

We investigate the photoluminescence (PL) and amplified spontaneous emission (ASE) of the quasi-2D emitter ($CsPbBr_3$ with 80% butylammonium bromide), and its 3D analogous formed by thermal removing the organic spacer (Fig 1a). Although the PL from the quasi-2D films is much brighter at low excitation power (Fig 1a), the ASE thresholds (600 μJ $cm^{-2}$) of the quasi-2D materials tend to be higher than the 3D counterparts (130 μJ $cm^{-2}$). This counter-intuitive behaviour is investigated through time-resolved photophysical studies, which reveal the emission in quasi-2D perovskite originated from the excitonic emission (Fig 1b). This accounts for its superior PL at low fluence, as the excitonic emission is efficient at low excited-state densities (Fig 1c). However, the 2$^{nd}$ order exciton–exciton annihilation of quasi-2D perovskite starts to take over the exciton dynamics at a low exciton density (<$10^{16}$ $cm^{-3}$), resulting in a low radiative efficiency at around transparency carrier density ($10^{18}$ $cm^{-3}$). Hence, to achieve the ASE in quasi-2D film, a much higher excitation fluence is necessary to increase the photon density in this low radiative efficiency regime. In contrast, the 2$^{nd}$ order free-carrier radiative recombination in 3D film leads to a high radiative efficiency steadily increasing to the transparency carrier density, which explains its lower ASE threshold. Through further examining the ASE thresholds of a series of quasi-2D perovskites with different 2D spacer content and type (i.e. 2-phenylethylammonium bromide and 1-naphthylmethylamine bromide), we highlight that quasi-2D perovskite gain materials should target fast free carrier recombination by engineering the thickness and size of QW, but not maximum PL quantum yields under low power excitation.

## REFERENCES

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## FIGURES

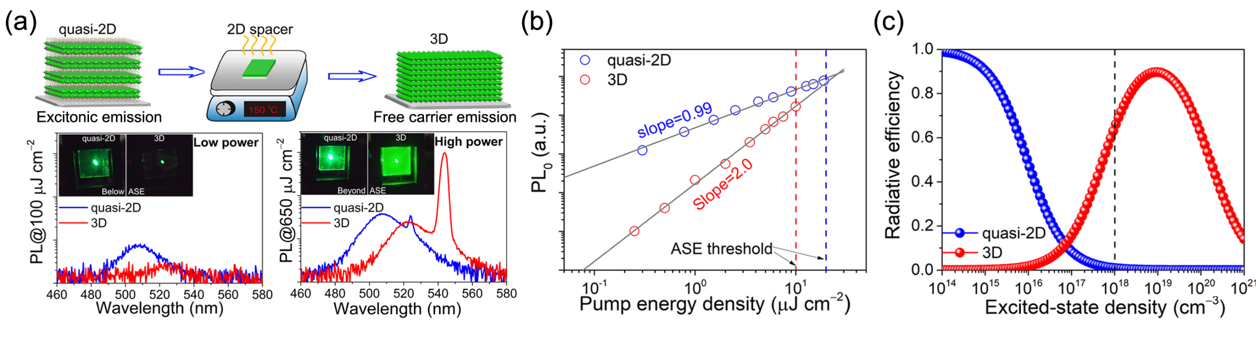


**Figure 1:** (a) Transform from the quasi-2D into a 3D perovskite film through an annealing step (top). PL/ASE spectra of quasi-2D and 3D perovskite films (bottom). (b) Plots of $PL_0$ versus the pump energy densities. (c) Calculated radiative efficiency. The dash line indicates the transparency carrier density.