

Two-Dimensional Electron–Hole Plasma in Colloidal Quantum Shells Enables Integrated Lasing Continuously Tunable in the Red Spectrum

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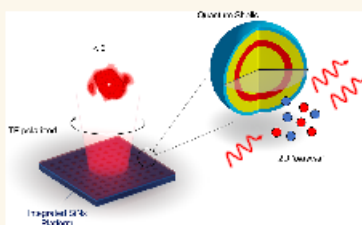
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ABSTRACT: Combining integrated optical platforms with solution-processable materials offers a clear path toward miniaturized and robust light sources, including lasers. A limiting aspect for red-emitting materials remains the drop in efficiency at high excitation density due to non-radiative quenching pathways, such as Auger recombination. Next to this, lasers based on such materials remain ill characterized, leaving questions about their ultimate performance. Here, we show that colloidal quantum shells (QShs) offer a viable solution for a processable material platform to circumvent these issues. We first show that optical gain in QShs is mediated by a 2D plasma state of unbound electron–hole pairs, opposed to bound excitons, which gives rise to broad-band and sizable gain across the full red spectrum with record gain lifetimes and a low threshold. Moreover, at high excitation density, the emission efficiency of the plasma state does not quench, a feat we can attribute to an increased radiative recombination rate. Finally, QShs are integrated on a silicon nitride platform, enabling high spectral contrast, surface emitting, and TE-polarized lasers with ultranarrow beam divergence across the entire red spectrum from a small surface area. Our results indicate QSh materials are an excellent materials platform to realize highly performant and compact on-chip light sources.

KEYWORDS: quantum shell, nanocrystal, optical gain, lasing, integrated photonics, photonic crystal



INTRODUCTION

With the advent of laser projection, augmented or virtual (AR/VR) reality headsets, and quantum communication, an increased demand for small footprint, bright, and highly collimated visible light sources is emerging. Moreover, such sources should be matched to very specific spectral bands, e.g., 620–640 nm to obtain pure red. Combining solution processable semiconductor gain materials with the diversity of scalable and miniaturization-driven integrated photonics platforms available today presents a promising route forward. In particular, the use of colloidal nanomaterials has pioneered this hybrid approach, showing optically pumped on-chip lasing, both with in and out-of-plane emission,^{1–3} and even demonstrations of electrically driven amplification.⁴ Such results are proof of a continued pursuit of more effective optical gain metrics and materials, resulting in the development

of a diverse range of nanocrystals with varying elemental compositions⁵ and structural configurations, e.g., by incorporating semiconductor shells to exploit charge separation,^{6,7} or by changing the geometry from 0D to 2D, leading to different photophysical mechanisms for light amplification.^{8,9} To understand what is lacking today, we can categorize these materials based on three nonlinear optical material parameters: the inverted-state (or gain –) lifetime, τ_G , the threshold carrier

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