

Biphasic interface-regulated ultrastable narrow-linewidth colloidal quantum well liquid lasers

Rui Duan^{1,2}, Bei Xu¹ (✉), Yuan Wang², Lin Wang³, Guodan Wei², Hao Wang¹, Kailei Lu¹, Yanli Shi¹, Xuyong Yang³, Yan Wang⁴ (✉), Jianqi Qi¹ (✉), Handong Sun² (✉)

¹ College of Physics, Sichuan University, Chengdu 610000, China

² Macao Centre for Research and Development in Advanced Materials, Institute of Applied Physics and Materials Engineering, University of Macau, Taipa, Macao 999078, China

³ Key Laboratory of Advanced Display and System Applications of Ministry of Education, Shanghai University, Shanghai 200072, China

⁴ School of Electronics and Information, Academy for Quantum Science and Technology, Zhengzhou University of Light Industry, Zhengzhou 450001, China

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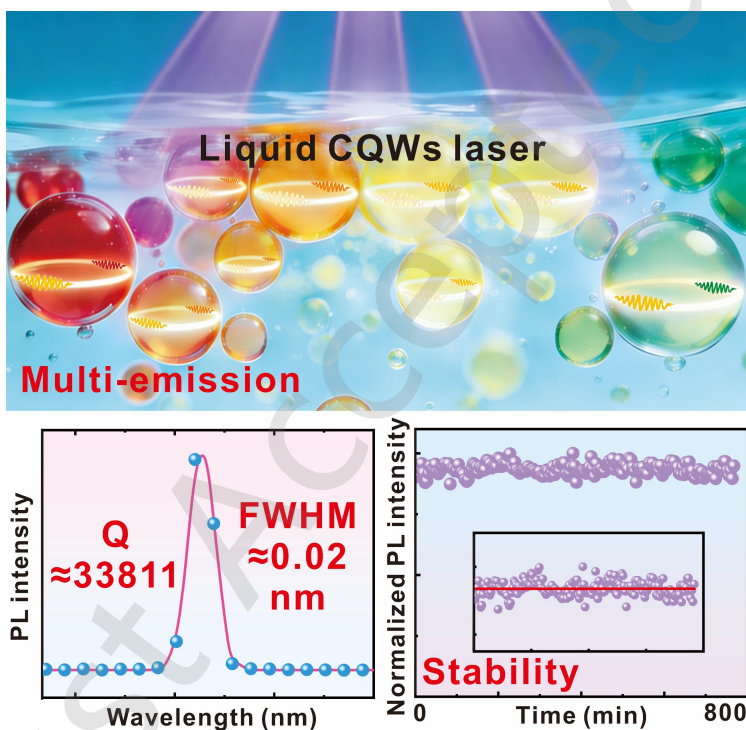
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¹ College of Physics, Sichuan University, Chengdu 610000, China

² Macao Centre for Research and Development in Advanced Materials, Institute of Applied Physics and Materials Engineering, University of Macau, Taipa, Macao S.A.R. 999078, China

³ Key Laboratory of Advanced Display and System Applications of Ministry of Education, Shanghai University, Shanghai 200072, China

⁴ School of Electronics and Information, Academy for Quantum Science and Technology, Zhengzhou University of Light Industry, Zhengzhou 450001, China



Liquid CQWs micro lasers with multi-color emission, ultra-high Q factor, and long-term stability.

Biphasic interface-regulated ultrastable narrow-linewidth colloidal quantum well liquid lasers

Rui Duan^{1,2}, Bei Xu ¹ , Yuan Wang², Lin Wang³, Guodan Wei², Hao Wang¹, Kailei Lu¹, Yanli Shi¹, Xuyong Yang³, Yan Wang ⁴ , Jianqi Qi ¹ , and Handong Sun ² 

¹ College of Physics, Sichuan University, Chengdu 610000, China

² Macao Centre for Research and Development in Advanced Materials, Institute of Applied Physics and Materials Engineering, University of Macau, Taipa, Macao 999078, China

³ Key Laboratory of Advanced Display and System Applications of Ministry of Education, Shanghai University, Shanghai 200072, China

⁴ School of Electronics and Information, Academy for Quantum Science and Technology, Zhengzhou University of Light Industry, Zhengzhou 450001, China

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 Address correspondence to Bei Xu, xybei@stu.scu.edu.cn; Yan Wang, ywang@zzuli.edu.cn; Jianqi Qi, qijianqi@scu.edu.cn; Handong Sun, hdsun@um.edu.mo

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ABSTRACT: Liquid nanocrystal microlasers are promising miniaturized coherent light sources, yet their development remains constrained by the difficulty of simultaneously preserving colloidal gain media, stabilizing liquid-cavity morphology, and maintaining low-loss optical feedback. Here, we report, to the best of our knowledge, the first colloidal quantum well (CQW)-based biphasic liquid microsphere laser and establish biphasic interfacial engineering as an active strategy for liquid nanocrystal lasing. Green-emitting CdSe/CdSe_{1-x}S_x core/crown CQWs and red-emitting CdSe/Cd_{1-x}Zn_xS core/shell CQWs are incorporated into glycerol/toluene biphasic microspheres to construct whispering gallery mode (WGM) microlasers with chromaticity-tunable emission. The glycerol/toluene interface functions as a multifunctional regulatory boundary that stabilizes the spherical microcavity, preserves the CQW microenvironment, and provides refractive-index contrast for WGM confinement. Consequently, the green- and red-emitting CQW microsphere lasers show low-threshold, spectrally pure lasing with a linewidth of ~0.02 nm, a polarization degree of 0.923, and a quality factor up to 3.27×10^4 . The red-emitting device exhibits negligible decay after 800 min at 10-fold threshold pumping. Furthermore, co-confinement of red and green CQWs within a single microsphere enables continuously tunable dual-color lasing. These results establish biphasic liquid-liquid interfaces as active mediators of nanocrystal photophysics, cavity stability, and optical feedback for stable and chromaticity-tunable liquid microlasers.

KEYWORDS: colloidal quantum wells, liquid microlasers, high stability, narrow linewidth, multicolor laser

1 Introduction

Miniaturized coherent light sources that combine high spectral purity, long operational stability, and tunable spectral output are increasingly important for integrated photonics, optical communications, display technologies, and microscale sensing [1-7]. Their linewidth, output stability, and wavelength tunability directly determine the performance limits of photonic devices, ranging from on-chip optical integration to precision optical readout and color-selective emission. In this context, liquid lasers are attractive because liquid gain media offer efficient heat dissipation, high optical gain, solution processability, and facile wavelength tuning [8,9]. Recent advances in colloidal QD lasers have demonstrated tunable and robust lasing, highlighting the potential of colloidal nanocrystals as solution-processable gain media for liquid laser applications [10]. Traditional dye-based liquid lasers have been widely investigated owing to the broad spectral tunability and mature photophysics of organic gain media [4,11]. However, their practical application remains

constrained by intrinsic molecular instability. Under sustained optical excitation, organic dyes are prone to photobleaching, resulting in rapid emission decay and degraded operational reliability [12]. These limitations motivate the development of alternative solution-processable gain materials that retain the tunability and processing advantages of liquid media while offering substantially improved photochemical stability.

Colloidal quantum wells (CQWs), also known as colloidal nanoplatelets, are quasi-two-dimensional semiconductor nanocrystals with atomically controlled thickness, strong quantum confinement, and large excitonic oscillator strength. These characteristics make them attractive gain media for solution-processable and liquid-phase microlasers [13-17]. Compared with traditional dyes, CQWs exhibit superior photochemical robustness, as their semiconductor crystal framework can effectively resist photoinduced damage and significantly suppress photobleaching [18-22]. In particular, CdSe/CdS core-shell CQWs have also shown highly photostable multiphoton-pumped lasing in solution, with an ultralow two-photon

lasing threshold of $1.2 \text{ mJ}\cdot\text{cm}^{-2}$ [15]. Moreover, CQWs possess optimized core-shell passivation structures that greatly suppress non-radiative recombination, enabling high photoluminescence quantum yields for efficient light conversion. They can also be well dispersed in organic solvents and exhibit long-term stable lasing performance with excellent photostability [23-25]. These features make CQWs highly promising as liquid-phase gain media. Recent developments in colloidal QD lasers have demonstrated full-color lasing with low thresholds, prolonged operation, and multimilliwatt output, further highlighting the potential of colloidal nanocrystals for high-performance laser applications [26]. However, replacing organic dyes with nanocrystal gain materials alone does not fully resolve the broader challenge of liquid microlaser design, because colloidal dispersion, cavity morphology, and low-loss optical feedback must be simultaneously preserved during prolonged operation [27].

Beyond high-performance gain media, the design of an appropriate optical cavity is equally crucial for achieving efficient and stable liquid microlasers [28-31]. Optical microcavities can confine photons in small mode volumes through high-quality (Q) resonances, thereby enhancing light-matter interaction, reducing lasing thresholds, and improving spectral purity. Among different resonator geometries, spherical microcavities are particularly attractive because their smooth closed boundaries support whispering-gallery-mode (WGM) resonances through total internal reflection, providing low-loss circulating optical feedback [32-36]. Such microsphere-type WGM resonators are especially compatible with colloidal nanocrystal and CQW gain media, as the resonant optical field can spatially overlap with the emitters while maintaining narrow spectral feedback. Recent progress in microfluidic QD lasers has further demonstrated reliable lasing and on-chip optical functionality using liquid-compatible optofluidic platforms [37]. Nevertheless, CQW-based liquid microsphere lasers impose a more stringent set of coupled requirements than solid or dried self-assembled resonators. The gain medium, liquid environment, and optical boundary are no longer independent. An unstable liquid cavity may deform or evaporate, increasing scattering and radiation losses; an incompatible liquid phase may induce CQW aggregation or emission degradation, and insufficient refractive-index contrast may weaken WGM confinement and reduce resonant feedback efficiency. Therefore, the key challenge is not merely to introduce CQWs into a liquid microcavity, but to engineer the liquid environment so that colloidal dispersion, cavity morphology, and optical boundary conditions are simultaneously stabilized during lasing operation.

Here, we report a CQW-based liquid microsphere laser that addresses this coupled challenge through a glycerol/toluene biphasic architecture and, more importantly, propose a biphasic interfacial co-regulation mechanism. Unlike conventional liquid-laser systems, in which the liquid phase mainly serves as a passive carrier for the gain medium, the liquid-liquid interface in our system functions as an active regulatory unit that simultaneously controls droplet morphology, the local microenvironment of CQWs, and the boundary conditions governing WGM confinement. In this design, glycerol, a high-viscosity and low-volatility solvent, serves as the outer continuous phase to suppress droplet deformation and evaporation, thereby stabilizing the spherical

microcavity geometry. Toluene, which exhibits excellent compatibility with CQWs, acts as the inner dispersing medium to preserve uniform nanocrystal dispersion and emissive integrity. At the same time, the refractive-index contrast between the two phases is optimized to strengthen WGM confinement and reduce optical leakage. As a result, the biphasic interface unifies three functions within a single microcavity platform: morphological stabilization, gain preservation, and efficient optical feedback. This mechanism enables robust coupling between nanocrystal gain media and a high-Q liquid microcavity, thereby suppressing deformation-induced optical loss while sustaining strong resonant feedback. Overall, this work reveals that biphasic interfaces can mediate the synergistic coupling among nanocrystal photophysics, cavity stability, and optical feedback, thereby providing a mechanism-based design principle for highly stable, chromaticity-tunable liquid microlasers.

2 Results and discussion

2.1 Preparation and characterization of liquid microsphere lasers based on CQWs

We synthesized two types of CQWs materials using the high-temperature hot injection method: green-emitting CdSe/CdSe_{1-x}S_x core-crown [23] (denoted as CdSe/CdSeS C/C) structure and red-emitting CdSe/Cd_{1-x}Zn_xS core-shell (denoted as CdSe/CdZnS C/S) structure. The former achieves bandgap modulation and spectral tuning through lateral coating of a crown layer, while the latter employs a three-dimensional shell to enhance optical stability and improve luminescence efficiency. By precisely controlling key process parameters, including precursor injection rate, reaction temperature, and holding time, we achieved controlled preparation of materials in terms of size, composition, and crystal structure, ensuring excellent optical stability and high photoluminescence quantum yield. These nanocrystals serve as the gain medium not only for efficient lasing, but also for probing how nanocrystal photophysics can be coupled to liquid microcavity optics through interfacial engineering.

On this basis, we further constructed glycerol/toluene biphasic liquid microspheres to establish a liquid microcavity platform in which CQW gain preservation, droplet morphological stability, and WGM boundary conditions could be co-regulated. Unlike conventional liquid-laser preparation strategies that mainly focus on dispersing the gain medium, our design uses the biphasic liquid-liquid interface as an active structural and optical regulator. By systematically adjusting parameters such as stirring rate, the volume ratio of the two phases, CQWs doping concentration, and the sequence of reagent addition, combined with interfacial tension control and self-assembly techniques, we obtained uniformly dispersed and size-uniform liquid microspheres (Figure 1a). Fluorescence microscopy images (Figure 1b) show that the toluene microspheres loaded with CQWs exhibit regular spherical shapes in the glycerol continuous phase, with no significant aggregation, adhesion, or sedimentation. Under excitation, they emit bright green fluorescence. To ensure consistency and comparability, all fluorescence images were acquired under the same imaging conditions, including the excitation wavelength, objective, exposure time, and detector gain, without additional adjustment of brightness or contrast after acquisition. The observed green fluorescence originates from the characteristic photoluminescence of the incorporated CQWs, whose

emission characteristics determine the fluorescence color of the microspheres. This highly dispersed microsphere structure is crucial for ensuring the consistency of WGM resonant modes, reducing mode distortion, and achieving stable and repeatable laser output.

Glycerol, as the high-viscosity continuous phase, has a refractive index of 1.47. The refractive index of the toluene-CQWs microspheres was adjusted to 1.59 through optimization of the doping concentration, meeting the refractive index matching condition required for WGM resonances. This laid a solid foundation for efficient localization of the optical field and stable resonance. To verify the intrinsic structure and chemical composition of the materials, we performed full-spectrum and high-resolution fine spectral characterization of the green-emitting CQWs using X-ray photoelectron spectroscopy (XPS). The full XPS spectrum in Figure 1c clearly shows characteristic peaks for elements such as Cd, Se, and S, with no obvious impurity elements, confirming the high purity of the target product. In Figure S1, the peak centered at 284.8 eV in the C 1s spectrum is attributed to C-C and C-H bonds originating from surface organic ligands, which are widely recognized to facilitate the dispersion of CQWs in organic solutions. Figures 1d-f display high-resolution fine spectra for Cd 3d, Se 3d, and S 2p orbitals. After peak fitting, the Cd 3d spectrum shows characteristic peaks at 405.4 eV, 412.2 eV, 404.9 eV, and 411.9 eV, corresponding to the $3d_{5/2}$ and $3d_{3/2}$ spin-orbit split peaks of Cd^{2+} , which correspond to Cd-S and Cd-Se bonding environments. Owing to the higher electronegativity of S relative to Se, the binding energies of Cd-S species shift 0.3–0.5 eV toward higher values compared with Cd-Se counterparts, demonstrating the coexistence of Cd-Se and Cd-S bonds within the as-prepared CdSe/CdSeS C/C nanostructure. In the high-resolution Se 3d spectrum, the characteristic peaks at 53.9 eV and 54.8 eV are assigned to the spin-orbit split $3d_{5/2}$ and $3d_{3/2}$ signals of lattice Se^{2-} , respectively. For the S 2p spectrum, the two peaks at 160.2 eV and 161.6 eV correspond to the $2p_{3/2}$ and $2p_{1/2}$ orbitals of S^{2-} , and the energy separation of this doublet matches the typical spin-splitting feature of sulfide anions. Combined with the Cd 3d results above, these spectra verify the stable divalent state of Cd species and the coexistence of Cd-Se and Cd-S bonds in the samples, confirming that the as-synthesized CdSe/CdSeS C/C CQWs contain both selenide and sulfide chemical environments.

To further verify the morphology, elemental distribution, and surface chemistry of the as-prepared CQWs, complementary characterizations were performed. Scanning electron microscopy (SEM) images show that the green-emitting CdSe/CdSeS C/C CQWs present uniform one-dimensional nanostructures with excellent dispersibility (Figure S2), whereas red-emitting CdSe/ $\text{Cd}_{1-x}\text{Zn}_x\text{S}$ CQWs exhibit homogeneous morphology and narrow size distribution (Figure S3). Statistical analyses based on the corresponding TEM images further reveal relatively narrow distributions of the long-axis length and short-axis width for both types of CQWs (Figures S8 and S9), indicative of their good dimensional uniformity and high quality. Energy-dispersive X-ray spectroscopy (EDS) elemental mapping confirms the omnipresence of Cd, Se, and S across the whole region of green CQWs (Figure S4), as well as Cd, Se, Zn and S in red CQWs (Figure S5), demonstrating homogeneous elemental composition throughout individual nanostructures. These

results collectively illustrate that both green- and red-emitting CQWs satisfy the requirements of compositional uniformity, structural integrity and colloidal stability for integration into biphasic liquid microcavities. More importantly, these observations underpin the core mechanism proposed in this work: high-quality CQW gain media and the biphasic liquid-liquid interface are not isolated units but synergistic components, which jointly contribute to stable optical gain, intact cavity geometry and efficient WGM mode confinement within a single liquid microsphere laser.

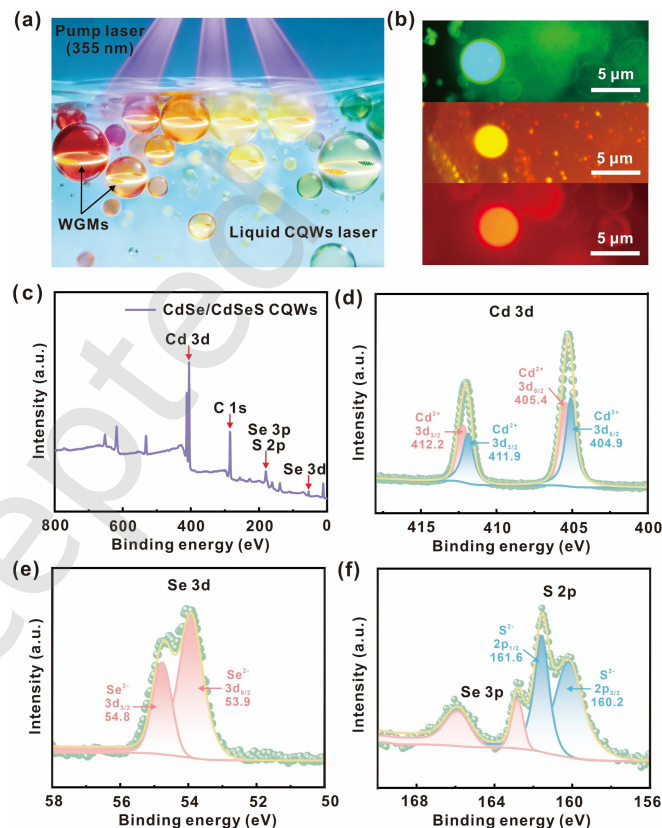


Figure 1. Preparation and material characterization of liquid quantum well microsphere lasers. (a) Schematic diagram of the whispering-gallery mode (WGM) resonance of the liquid microsphere laser. (b) Photos of different liquid microsphere lasers. (c) X-ray photoelectron spectroscopy (XPS) full spectrum of green-emitting colloidal quantum wells (CQWs). (d–f) XPS fine spectra of green-emitting CQWs, with peak fitting results for the chemical states of Cd 3d, Se 3d, and S 2p orbitals. The scale bar in (b) is 5 μm.

2.2 Laser characteristics and green-emitting CQW-based biphasic liquid microsphere lasers

To explore the optical characteristics of the liquid CQWs microsphere cavity, we numerically simulated and verified the resonance mechanism of the WGM. Figure 2a shows the electric field distribution cross-section of a 39-μm-diameter microsphere cavity, where the strong localization effect of the optical field at the microsphere interface can be clearly observed. The light is confined to the interface gain layer via total internal reflection and only weakly leaks as an evanescent wave into the glycerol continuous phase. The significant concentration of the interface electric field confirms the formation of low-loss resonant feedback, providing the core support for optical amplification in the microsphere laser. Based on this optical field confinement, we further characterized the optical response of the gain material to assess its spectral matching with the resonant modes. Figure 2b shows the absorption and PL spectra of the green-emitting

CdSe/CdSeS C/C CQWs. The absorption spectrum exhibits a strong absorption peak at 511.5 nm, corresponding to the heavy-hole (*hh*) interband transition. In the high-energy region, characteristics of light-hole (*lh*) transitions are observed, reflecting the electronic state structure of quasi-two-dimensional quantum wells [24, 38]. Additionally, the broad photoluminescence (PL) emission envelope of the CQWs spectrally encompasses multiple WGM resonant peaks of the microsphere cavity, with the peak gain wavelength (514 nm) closely matching the fundamental WGM mode family. This spectral matching, combined with the high photoluminescence quantum yield (~99%) and a narrow gain bandwidth (8 nm full-width at half-maximum (FWHM)), establishes the foundation for low-threshold lasing through efficient stimulated emission coupling into the high-Q WGMs.

Once the spectral compatibility was clarified, we investigated the lasing behavior of the microsphere laser through pump power-dependent measurements. Figure 2c shows the pump power dependence of multi-mode lasing. Under pulsed optical pumping, the integrated PL intensity exhibits a characteristic threshold behavior. Below a pump fluence of 59 $\mu\text{J}/\text{cm}^2$, the emission intensity increases linearly with pump power, which is consistent with spontaneous emission dominance; upon exceeding the threshold, the intensity rises superlinearly accompanied by the emergence of multiple spectrally narrow lasing peaks, unambiguously indicating the onset of stimulated emission. This threshold reflects the establishment of sufficient optical gain under conditions where the biphasic microcavity already provides stable resonant feedback and low optical loss. In other words, lasing in this system is not solely determined by the CQW gain medium or the spherical geometry alone, but by the cooperative balance among gain buildup, cavity stability, and interface-defined optical confinement.

To further clarify the lasing mechanism, we analyzed the resonance modes of the microcavity in detail. In the spherical microsphere, light is efficiently confined through total internal reflection at the interface between the toluene/CQW inner phase ($n_1 = 1.59$) and the surrounding glycerol continuous phase ($n_2 = 1.47$), satisfying the total-internal-reflection condition of $n_1 > n_2$. Importantly, this refractive-index contrast is established and maintained by the biphasic architecture, underscoring that the liquid-liquid interface is a decisive regulator of WGM formation rather than a passive phase boundary. By substituting the microsphere diameter ($D = 39 \mu\text{m}$) and the refractive indices into the WGM eigenfrequency equation (Equation S1), the theoretical lasing positions agree well with the experimental spectra (Figure 2d). High-resolution mode assignment shows that all lasing peaks can be attributed to first-order ($r = 1$) transverse magnetic (TM) and transverse electric (TE) modes, with angular mode numbers m ranging from 361 to 366 and wavelengths from 516 to 524 nm. Detailed parameters of the TM and TE modes, including theoretical and experimental wavelengths, are summarized in Table S1. The mode spacing is in excellent agreement with the theoretical free spectral range (FSR): the calculated FSR is 1.40 nm, while the experimental spacings are 1.40 nm for TM modes and 1.39 nm for TE modes. These results directly confirm that the observed lasing originates from stable high-Q WGM resonances, where the optical feedback is governed by the spherical cavity geometry and the liquid-liquid interfacial boundary

conditions, while the CQW gain medium provides the necessary population inversion for stimulated emission amplification.

To broaden the application potential of the device, we further explored single-mode lasing in smaller microsphere cavities. Figure 2e shows the single-mode lasing behavior of a reduced-size microsphere, with a threshold of 87.4 $\mu\text{J}/\text{cm}^2$. Below the threshold, only broad spontaneous emission is observed; once the pump fluence exceeds the threshold, the spectrum becomes dominated by a single narrow peak. This behavior originates from the reduced cavity size, which increases the FSR and therefore limits the number of supported modes within the gain bandwidth, while gain-saturated mode competition further enhances the selectivity for the lowest-threshold mode. From the mechanistic perspective, this size-dependent transition from multimode to single-mode lasing demonstrates that once the biphasic interface stabilizes the cavity and preserves gain quality, the output modal characteristics can be predictably tuned through cavity-size-dependent optical boundary conditions. Figure 2f presents a high-resolution enlargement of the single-mode peak, showing a linewidth of only 0.02 nm. The inset compares the emission images below and above threshold: at 5.2 $\mu\text{J}/\text{cm}^2$, only diffuse weak green fluorescence is observed, whereas at 93.4 $\mu\text{J}/\text{cm}^2$ a bright and sharply defined lasing pattern appears, clearly marking the lasing transition.

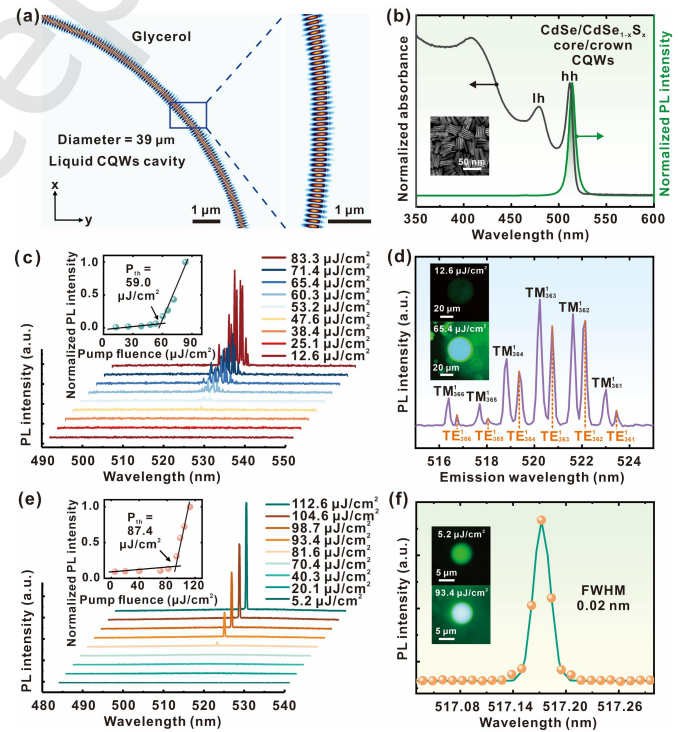


Figure 2. Laser characteristics of green-emitting CQW microsphere lasers. **(a)** Simulation results of the electric field distribution in the WGM resonator. **(b)** Absorption and photoluminescence (PL) spectra of green CdSe/CdSeS core/crown (C/C) CQWs, with the inset showing the scanning electron microscopy (SEM) image. **(c)** Pump power-dependent PL intensity of the multimode microsphere laser, with the inset showing the laser threshold fitting results. **(d)** Multimode laser emission spectrum, with the peak positions and mode numbers of the main modes labeled according to the WGM mode formula. **(e)** Pump power-dependent PL intensity of the single-mode microsphere laser, with the inset showing its low-threshold characteristics. **(f)** Magnified spectrum of a single laser peak from (e), with the inset comparing the emission photos of the microsphere before and after reaching the lasing threshold. The scale bars in (d) and (f) are 20 μm and 5 μm , respectively.

CdSe/CdSeS C/C CQWs, as quasi-two-dimensional gain

media, possess high excitonic oscillator strength and a narrow gain bandwidth arising from quantum confinement-induced discrete density of states. These properties allow efficient coupling to WGM resonances. Meanwhile, the WGM microcavity provides low-loss optical feedback, and the evanescent field overlaps with the CQW gain region to support effective stimulated-emission amplification. Multimode lasing arises in larger cavities because multiple closely spaced WGMs fall within the gain bandwidth, whereas single-mode lasing is achieved by reducing the cavity size to enlarge the FSR, reduce the mode volume, and enhance mode competition. Notably, the single-mode threshold ($\sim 87 \mu\text{J}/\text{cm}^2$) is moderately higher than the multimode threshold ($\sim 59 \mu\text{J}/\text{cm}^2$), reflecting the reduced gain volume in the smaller cavity, yet both remain in the sub-hundred- $\mu\text{J}/\text{cm}^2$ regime, highlighting the high optical gain of CQWs and the low-loss characteristics of the biphasic WGM microcavity. More importantly, these results show that the laser characteristics are governed by a unified mechanism in which the biphasic interfacial architecture defines the cavity morphology and optical confinement conditions, while preserving the intrinsic high gain quality of the CQWs. The linewidth evolution with pump fluence further confirms the transition from spontaneous emission to stimulated emission, with obvious linewidth narrowing above threshold for both single-mode and multimode lasing (Figure S6). Therefore, the green-emitting liquid CQW microsphere laser serves not only as a high-performance coherent light source, but also as direct evidence that the proposed biphasic interfacial co-regulation mechanism can translate into stable, low-threshold, and mode-tunable lasing behavior.

2.3 Laser characteristics, polarization response, and operational stability of red-emitting CQW-based biphasic liquid microsphere lasers

Compared with green-emitting CQWs, red-emitting CdSe/CdZnS C/S CQWs extend the emission wavelength into the visible red region through a core/shell heterostructure. This design not only addresses the demand for long-wavelength coherent sources in fields such as biological imaging and display technologies, but also preserves the excellent optical properties of CQWs, including high quantum yield and narrow emission bandwidth. More importantly, the red-emitting CQWs provide an alternative gain medium platform for examining whether the proposed biphasic interfacial co-regulation mechanism can be generalized to different spectral regions and quantum-well heterostructure designs. In this sense, the red-emitting liquid microsphere laser is not merely a wavelength extension of the green system, but a critical validation of the generality of the biphasic cavity-gain architecture.

Figure 3a shows the normalized absorption and PL spectra of the red-emitting CQWs. The absorption band arises from the *lh* and *hh* excitonic transitions, and the PL peak is centered at 640 nm with a FWHM of 24 nm, exhibiting the characteristic gain behavior in the red spectral region. This red-shifted yet well-defined gain profile provides suitable spectral overlap with the corresponding WGM resonances of the microsphere cavity, thereby validating the spectral universality of the gain-cavity matching mechanism established for the green-emitting system. Based on this spectral compatibility, we fabricated a single-mode red microsphere laser and systematically investigated its lasing behavior. Figure 3b

presents the PL spectra recorded under different pump fluences. When the pump fluence is below threshold, the emission is dominated by spontaneous radiative recombination; once the threshold is exceeded, sharp laser peaks emerge. The inset, showing the normalized PL intensity and the fitted threshold curve as a function of pump fluence, indicates that the threshold pump fluence is as low as $42.9 \mu\text{J}/\text{cm}^2$, reflecting the high gain efficiency of the red-emitting CQWs and the optimized single-mode cavity configuration. This low threshold further confirms that the biphasic microcavity architecture enables efficient gain-resonance coupling from the green to the red spectral region, underscoring the generality of the biphasic interfacial co-regulation mechanism.

A magnified view of a representative lasing peak is shown in Figure 3c, revealing an emission linewidth of $\sim 0.02 \text{ nm}$. The pump-fluence-dependent evolution of this peak exhibits a clear threshold and pronounced linewidth narrowing above threshold (Figure S7), confirming the onset of stable single-mode lasing. The corresponding Q factor reaches 3.27×10^4 , further demonstrating the outstanding optical feedback capability of the microcavity. The inset compares the emission morphology below and above threshold: below threshold ($5.2 \mu\text{J}/\text{cm}^2$), the emission appears diffuse, whereas above threshold ($93.4 \mu\text{J}/\text{cm}^2$), it evolves into a bright and sharply defined lasing pattern. Together, these results demonstrate that the biphasic liquid microcavity can support high-Q and low-loss lasing over different spectral windows, indicating that the interfacial co-regulation mechanism is spectrally universal and architecturally robust.

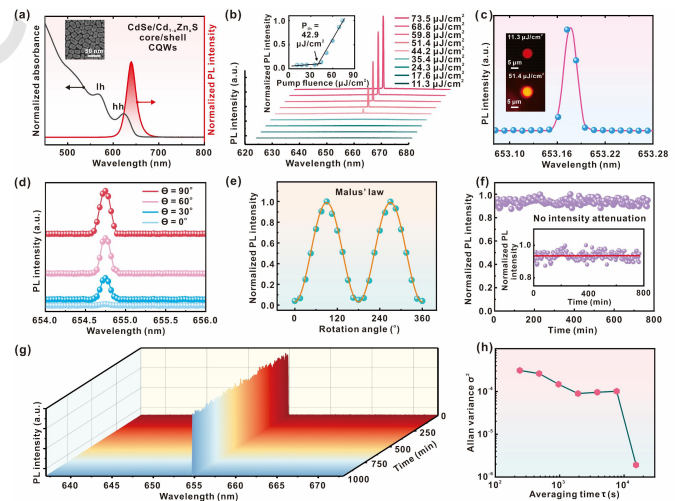


Figure 3. Laser characteristics, polarization response, and long-term stability of red-emitting CQW microsphere lasers. (a) Absorption and PL spectra of red CdSe/CdZnS core/shell (C/S) CQWs, with the inset showing the SEM image. (b) Pump power-dependent PL intensity of the red single-mode microsphere laser, with the inset showing the laser threshold. (c) Magnified spectrum of a single laser peak from (b), with the inset showing the emission photos before and after reaching the lasing threshold. (d) PL emission spectra at different polarization angles. (e) Polarization data fitting curve based on Malus' law. (f) Laser output intensity stability curve under 800 min of continuous pumping. (g) Overlay of all raw PL spectra recorded during the stability test. (h) Allan deviation calculated from the data in (g). The scale bar in (c) is $5 \mu\text{m}$.

Polarization response is another important indicator of the optical performance of microsphere lasers. Figure 3d shows the PL spectra measured at different polarization angles ($\theta = 0^\circ, 30^\circ, 60^\circ$, and 90°). The laser intensity varies strongly with the analyzer angle, reaching a maximum at $\theta = 90^\circ$ and being nearly suppressed at $\theta = 0^\circ$, indicating

pronounced linear polarization characteristics. This polarization-selective output reflects the well-defined modal characteristics of the WGM cavity and further indicates that the interfacially stabilized microcavity can sustain a dominant polarization eigenstate with high reproducibility. Further fitting of the polarization-dependent intensity according to Malus' law (Figure 3e) yields an excellent cosine-squared dependence, confirming that the red microsphere laser behaves as an effectively linearly polarized source. The degree of polarization (DoP) is calculated to be 0.923. The high DoP indicates that mode competition or slight cavity shape anisotropy preferentially selects a single polarization family (TE or TM), suppressing the orthogonal counterpart. From the mechanistic perspective, this result indicates that the biphasic interface not only stabilizes the cavity geometry and gain environment, but also maintains well-defined polarization eigenstates through the WGM mode anisotropy and interfacial optical boundary conditions.

Operational stability is a key factor in evaluating the practical applicability of microlasers. We therefore monitored the laser output continuously for 800 min under pulsed pumping at ten times the threshold ($\sim 430 \mu\text{J}/\text{cm}^2$). As shown in Figure 3f, the normalized lasing intensity exhibits negligible degradation throughout the test, demonstrating excellent optical stability. The inset presents a linear fit of the time-dependent intensity, with an extremely small slope, visually confirming the negligible fluctuation of the laser output. Figure 3g shows the overlaid PL spectra collected during the stability measurement, where both the peak position and the intensity remain highly stable without obvious drift or attenuation. Quantitative analysis of the spectral time series was performed using Allan deviation (Figure 3h). The results show that, for averaging times $\tau < 104$ s, the intensity fluctuation σ_2 remains on the order of 10^{-4} , and approaches a minimum value of 1.9×10^{-6} at $\tau = 1.5 \times 10^4$ s, indicating white-noise-dominated behavior with minimal long-term drift. These results demonstrate that the long-term operational stability of the red-emitting liquid microsphere laser does not arise solely from the intrinsic photostability of the CQWs, but from the cooperative stabilization of the cavity-gain system enabled by the biphasic interface. In other words, the liquid-liquid interface provides mechanical stability by suppressing droplet deformation, thereby preserving stable WGM boundary conditions, while simultaneously maintaining a benign local microenvironment that protects the CQWs from photodegradation and aggregation, thereby translating the proposed mechanism into highly durable lasing output. Taken together, the red-emitting system confirms that the biphasic interfacial co-regulation mechanism can support not only high-quality lasing and polarization-selective emission, but also exceptional long-term stability, providing strong evidence for its potential as a promising design strategy for liquid nanocrystal microlasers across multiple spectral regimes.

2.4 Tunable chromaticity in a single CQW-based biphasic liquid microsphere

Multicolor lasers have attracted considerable attention because of their tunable spectral properties, compact device architecture, and excellent beam quality, making them highly promising for integrated photonics, bioimaging, spectroscopic sensing, and advanced display technologies. However, the central challenge in multicolor

microlasers is not merely generating multiple wavelengths, but achieving stable and continuously tunable gain regulation within a single miniaturized cavity. Here, we exploit a single biphasic liquid microsphere as a shared WGM resonator and realize continuously tunable chromaticity by co-confining red- and green-emitting CQWs within the same optical cavity. Importantly, this strategy is not a simple spectral superposition of two emitters, but a cavity-mediated dual-gain regulation process in which two nanocrystal gain channels coexist within the same interfacially stabilized resonator, each supporting distinct WGM mode families while competing for pump energy in the spectral overlap region. By varying the ratio of red and green CQWs within a single microsphere while maintaining constant total CQW concentration, the relative gain contributions of the two channels can be continuously adjusted while maintaining stable cavity feedback. On this basis, five representative volume-fraction schemes were designed for the red/green CQW mixtures (green:red = 27:2, 21:6, 15:10, 9:14, and 3:18). As shown in Figures 4a-e, the single microsphere laser stably emits dual-wavelength output at ~ 515 nm and ~ 650 nm for all five ratios, without intermediate-wavelength emission or spurious multimode peaks. This result demonstrates that the biphasic microcavity can sustain simultaneous lasing of two spectrally distinct gain populations within one shared resonator, providing direct evidence for tunable dual-channel gain regulation. Furthermore, as shown in Figure S10, the red-to-green laser intensity ratio ($I_{\text{red}}/I_{\text{green}}$) exhibits a clear and monotonic increase with increasing red CQW volume fraction, while the total CQW concentration is kept constant. Specifically, $I_{\text{red}}/I_{\text{green}}$ increases from 0.12 at a red CQW volume fraction of 6.9% to 6.57 at 85.7%. This quantitative trend demonstrates that the relative intensities of the red and green lasing channels can be effectively regulated by varying the red/green CQW composition, thereby providing quantitative support for tunable spectral composition and chromaticity in the dual-wavelength microsphere laser.

The relative intensities of the two lasing peaks evolve continuously with the CQW composition ratio. As the fraction of green-emitting CQWs decreases, the red lasing peak gradually increases, while the green lasing peak correspondingly weakens. The output chromaticity can therefore be tuned smoothly from green-dominant to red-dominant emission. From the mechanistic perspective, this continuous evolution indicates that the biphasic interface not only stabilizes the cavity geometry and preserves the local microenvironment of both gain media, but also allows their relative gain contributions to be systematically adjusted within the same WGM cavity. The inset images in Figures 4a-e visually compare the microsphere emission below and above the lasing threshold for each composition ratio. The observed color evolution from pure green to pure red agrees well with the spectral results, clearly demonstrating the continuously tunable chromaticity of the single-microsphere laser. These observations further verify that the proposed biphasic interfacial co-regulation mechanism can be extended from single-color lasing to tunable dual-color output.

To quantitatively evaluate the chromaticity tunability of the microsphere laser, the emission spectra of the five CQW-ratio samples were converted into the CIE 1931 chromaticity coordinates, and the results are shown in Figure 4f. For comparison, the chromaticity coordinates of

pure green and pure red single-microsphere lasers were also included as reference points. The experimental coordinates are uniformly distributed within the effective color gamut and follow an approximately linear trajectory between the pure green and pure red reference points. This nearly linear chromaticity evolution indicates that the two gain channels operate relatively independently within the shared cavity, each supporting distinct WGM resonances with minimal cross-gain saturation. The linearity thus reflects predictable color mixing through simple compositional control, rather than unstable mode fluctuations or strong nonlinear gain competition. Therefore, the chromaticity tuning observed here is not only continuous, but also deterministically ratio-controlled and highly reproducible. This result quantitatively confirms that the chromaticity of the single microsphere laser can be precisely regulated through the red/green CQW ratio, while also highlighting the robustness and reliability of this tuning strategy. More broadly, it shows that biphasic liquid microcavities can serve as versatile platforms for multichannel nanocrystal lasing, opening opportunities for integrated multicolor photonic devices.

To further contextualize the performance of the present liquid CQW microsphere lasers, a comparison with representative state-of-the-art nanocrystal-based lasers is provided in Table S2. The red-emitting WGM laser in this work achieves a threshold of $42.9 \mu\text{J}/\text{cm}^2$ and a high Q factor of approximately 3.27×10^4 under nanosecond pumping, which is highly competitive among reported solution-processed colloidal nanocrystal WGM lasers. Combined with the tunable dual-color output, polarization-selective emission, and long-term operational stability demonstrated above, these results further indicate that the present biphasic CQW microsphere platform integrates high-quality lasing, exceptional stability, and controllable multichannel emission within a single liquid microresonator.

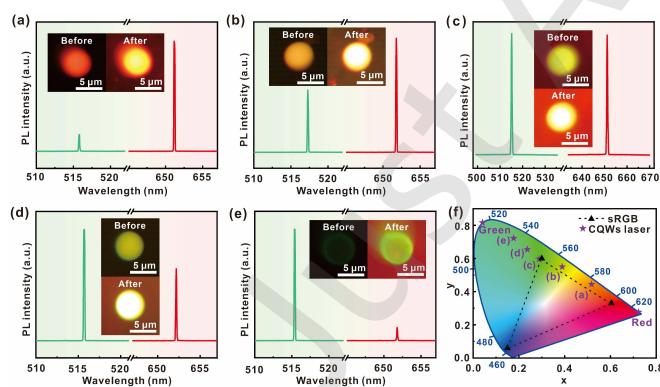


Figure 4. Multicolor laser sources. (a–e) PL spectra of a single microsphere laser with different ratios of red- and green-emitting CQWs. The inset in each panel shows the emission photos of the corresponding microsphere before and after reaching the lasing threshold. (f) Commission Internationale de l'Éclairage (CIE) 1931 chromaticity diagram, with the chromaticity coordinates of five single microspheres with different CQW ratios and the reference coordinates of pure green and pure red single microspheres. The scale bars in (a–e) are $5 \mu\text{m}$.

3 Conclusions

In conclusion, we have developed a CQW-based liquid microsphere laser using a glycerol/toluene biphasic system and, more importantly, established a biphasic interfacial co-regulation mechanism in which the liquid-liquid interface simultaneously defines the cavity geometry, protects the gain medium, and establishes optical confinement, unifying these functions within a single liquid microcavity. Rather than

treating the liquid environment merely as a passive carrier for the gain medium, this work demonstrates that the biphasic interface can function as an active regulator that mechanistically resolves the long-standing trade-off among gain-medium stability, cavity robustness, and optical confinement in conventional liquid microlasers. By controllably synthesizing high-optical-quality green- and red-emitting CQW gain materials and rationally matching the refractive indices of the glycerol outer phase and the toluene/CQW inner phase, we achieved robust coupling between nanocrystal gain media and a high- Q liquid microcavity, enabling low-loss and highly stable WGM lasing. The proposed liquid CQW microsphere laser exhibits a low lasing threshold of $\sim 43 \mu\text{J}/\text{cm}^2$ (red-emitting system), a spectrometer-limited linewidth of 0.02 nm corresponding to a quality factor of at least 3.27×10^4 , and a high degree of linear polarization (DoP = 0.923). Negligible degradation was observed after 800 min of continuous operation, confirming the exceptional operational stability enabled by the interfacially stabilized cavity-gain system. In addition, by co-encapsulating red- and green-emitting CQWs into a single microsphere, we established two gain channels sharing the same WGM microcavity, thereby enabling continuous tuning of dual-color lasing intensity and chromaticity through compositional regulation. This result indicates that the proposed biphasic interfacial mechanism not only stabilizes liquid nanocrystal lasing, but also enables tunable multichannel gain regulation within a shared optical resonator.

The present design principle may be extended by integrating blue-emitting CQWs, which may enable collectively tunable red, green, and blue gain channels within a single microsphere, toward full-visible-spectrum chromatic tuning. More broadly, the biphasic liquid microcavity strategy provides a versatile platform for highly stable and chromaticity-tunable microlasers, with potential applications in integrated displays, on-chip optical interconnects, and biochemical sensing. In addition, this interfacial design concept may be further expanded to multi-microsphere arrays and microfluidic integration, enabling high-throughput optical detection and signal processing. These findings therefore establish biphasic interfaces as a design principle for coupling nanocrystal photophysics, cavity stability, and optical feedback in liquid microlasers, and offer a promising route toward advanced miniaturized photonic devices.

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Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and/or the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

R.D.: Conceptualization, methodology, investigation, formal analysis, data curation, visualization, writing – original draft, writing – review & editing. B.X.: Methodology, investigation, writing – original draft. Yuan W.: Conceptualization, formal analysis, visualization. L.W.: Methodology, investigation, formal analysis. G.W.: Investigation, data analysis. H.W.: Investigation, validation. K.L.: Investigation, visualization. Y.S.: Investigation, validation. X.Y.: Investigation, software. Yan W.: Conceptualization, methodology, investigation, visualization, writing – review & editing. J.Q.: Supervision, investigation, resources, funding acquisition, writing – review & editing. H.S.: Conceptualization, investigation, supervision, project administration, funding acquisition, writing – review & editing. All the authors have approved the final manuscript.

Informed consent

Not applicable

Ethics statement

Not applicable

Use of AI statement

None

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Biphasic interface-regulated ultrastable narrow-linewidth colloidal quantum well liquid lasers

Rui Duan^{1,2}, Bei Xu ¹ , Yuan Wang², Lin Wang³, Guodan Wei², Hao Wang¹, Kailei Lu¹, Yanli Shi¹, Xuyong Yang³, Yan Wang ⁴ , Jianqi Qi ¹ , and Handong Sun ² 

¹ College of Physics, Sichuan University, Chengdu 610000, China

² Macao Centre for Research and Development in Advanced Materials, Institute of Applied Physics and Materials Engineering, University of Macau, Taipa, Macao 999078, China

³ Key Laboratory of Advanced Display and System Applications of Ministry of Education, Shanghai University, Shanghai 200072, China

⁴ School of Electronics and Information, Academy for Quantum Science and Technology, Zhengzhou University of Light Industry, Zhengzhou 450001, China

 Address correspondence to Bei Xu, xybei@stu.scu.edu.cn; Yan Wang, ywang@zzuli.edu.cn; Jianqi Qi, qijianqi@scu.edu.cn; Handong Sun, hdsun@um.edu.mo

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Experimental and Simulation Section

S1 Chemicals

All reagents were used as received without further purification. Technical grade 1-octadecene (ODE, 90%), oleic acid (OA, 90%), oleylamine (OLAm, 70%), cyclohexane (HPLC, $\geq 99.9\%$), selenium powder (trace metals basis $\geq 99.99\%$), Cadmium nitrate tetrahydrate ($\text{Cd}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$, 98%), cadmium acetate dihydrate ($\text{Cd}(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$, 98%), myristic acid ($\geq 99\%$), sodium hydroxide (97%), 1-octanethiol (98%), cadmium oxide (CdO , trace metals basis $\geq 99.99\%$), zinc acetate dihydrate ($\text{Zn}(\text{Ac})_2 \cdot 2\text{H}_2\text{O}$, trace metals basis $\geq 99.99\%$) were all purchased from Sigma-Aldrich. Hexane and methanol were purchased from Fisher Scientific.

S2 Synthesis of $\text{CdSe}/\text{Cd}_{1-x}\text{Zn}_x\text{S}$ core/shell CQWs

Core CdSe CQWs with a thickness of four monolayers were synthesized according to a well-established protocol reported previously and were used as seeds for the subsequent preparation of core/shell heterostructures [1]. Cadmium oleate (0.2 M) and zinc oleate (0.2 M) were prepared following a previously published protocol [2]. For the subsequent shell growth, the following precursor solutions were prepared: a cadmium precursor consisting of 2 mL of ODE and 2 mL of cadmium oleate; a zinc precursor consisting of 0.5 mL of ODE, 0.25 mL of oleylamine, and 0.7 mL of zinc oleate; and a sulfur precursor consisting of 4 mL of ODE and 84 μL of octanethiol.

For the synthesis of $\text{CdSe}/\text{Cd}_{1-x}\text{Zn}_x\text{S}$ core/shell CQWs, 8 mL of ODE was loaded into a 50 mL three-neck round-bottom flask and degassed at 60 $^{\circ}\text{C}$ for 30 min. The temperature was then raised to 300 $^{\circ}\text{C}$, and the solution was heated under a nitrogen atmosphere. At 150 $^{\circ}\text{C}$, 0.15 mL of cadmium precursor and sulfur precursor were introduced into the reaction solution at a rate of 7 mL h^{-1} . Subsequently, the CdSe core CQW dispersion was injected into the reaction mixture. The injected amount was equivalent to 2 mL of CdSe NPL dispersion with an optical density of 20 at the heavy-hole electron excitonic transition, measured in a 1 cm path-length quartz cuvette. After core injection, 2.6 mL of cadmium precursor and 2.6 mL of sulfur precursor were slowly added to the reaction flask at a rate of 7 mL h^{-1} to form a smooth CdS buffer layer. Then, 1 mL of zinc precursor and 1 mL of sulfur precursor were introduced at the same rate to grow the $\text{Cd}_{1-x}\text{Zn}_x\text{S}$ alloyed shell. After all precursors had been injected, the reaction mixture was annealed at 300 $^{\circ}\text{C}$ for an additional 20 min. The flask was then placed in an ice bath and cooled to room temperature. For purification of the core/shell CQWs, 10 mL of hexane and an appropriate amount of ethanol were added to the crude mixture, which was then centrifuged at 7000 rpm for 10 min. This purification step was repeated twice to remove excess precursors and quantum dot byproducts. The resulting precipitated CQWs were redispersed in cyclohexane and stored under ambient conditions for further characterization and measurements.

S3 Synthesis of CdSe/CdSeS core/crown CQWs

All reagents were used without further purification. The CdSe/CdSeS core/crown CQWs were synthesized as follows. First, 340 mg of $\text{Cd}(\text{myr})_2$, 24 mg of selenium powder, and 30 mL of ODE were loaded into a three-neck flask. The mixture was degassed at room temperature for 30 min, followed by further degassing at 100 $^{\circ}\text{C}$ for 10 min. Under a nitrogen atmosphere, the temperature was then increased to 240 $^{\circ}\text{C}$ at a heating rate of 10–12 $^{\circ}\text{C min}^{-1}$. When the reaction temperature reached 200–205 $^{\circ}\text{C}$, 120 mg of $\text{Cd}(\text{ace})_2 \cdot 2\text{H}_2\text{O}$ was added. After maintaining the reaction for 6 min following the addition of $\text{Cd}(\text{ace})_2 \cdot 2\text{H}_2\text{O}$, S-Sus-OA was injected at a rate of 18 mL h^{-1} for 5 min. The reaction mixture was then cooled to 160 $^{\circ}\text{C}$ using an air gun, after which 0.6 mL of OA was injected. Subsequently, the mixture was degassed at 110 $^{\circ}\text{C}$ for 10 min to remove H_2S . Once the temperature dropped below 70 $^{\circ}\text{C}$, 2 mL of OA and 5 mL of hexane were added. The final product was purified several times using ethanol and hexane.

S4 Measurement of Steady-state Photoluminescence (PL), Absorption

For steady-state PL/absorption measurement, a homebuilt optical setup was employed. The light sources were provided by a halogen tungsten lamp for absorption measurement and a He-Cd laser for PL measurement. Light signals were

resolved and recorded by a monochromator with a photomultiplier using a standard lock-in detection technique to improve the signal-to-noise ratio. The diffraction grating of monochromator was fixed at 600 grooves per mm and the measurement step was set as 0.2 nm.

S5 Scanning Electron Microscope (SEM), Transmission Electron Microscopy (TEM) and Energy Dispersive X-ray Spectroscopy (EDS) Characterization

SEM characterization was performed using FESEM JEOL JSM-7800F PRIME. TEM characterization was performed using JEOL JEM 1400 Electron Microscope. HAADF-STEM images were conducted by an aberration corrected JEOL ARM200F operating at 200 kV. The semiangle of the probe is ~ 30 mrad, and the collection angle is ~ 80 mrad to 280 mrad. Energy dispersive X-ray spectroscopy (EDS) analysis was carried out on the same aberration-corrected JEOL ARM200F microscope to identify the elemental composition and distribution.

S6 X-ray photoelectron spectroscopy (XPS)

XPS was performed using a Thermo Scientific ESCALAB 250Xi with Al K α radiation ($h\nu = 1486.6$ eV). Green CdSe/CdSeS CQWs were deposited on clean silicon substrates to form thin films for XPS measurements. Survey spectra were collected over 0–1000 eV, and high-resolution spectra of Cd 3d, Se 3d, and S 2p were acquired at a pass energy of 20 eV. Peak fitting was performed using XPS Peak 4.1 software to analyze chemical states and bonding environments.

Additional XPS measurements were conducted with a Kratos AXIS Ultra DLD using Al K α radiation ($h\nu = 1486.6$ eV), with raw data in .dset format. Green CQWs were deposited on silicon substrates, and survey spectra (0–1000 eV) and high-resolution spectra of Cd 3d, Se 3d, and S 2p were recorded at 20 eV pass energy. Survey spectra were calibrated using C 1s at 285 eV and high-resolution spectra using C 1s at 284.8 eV. Data fitting and chemical state analysis were performed with Advantage software.

For the high-resolution XPS spectra, peak fitting was performed using a Shirley-type background and Gaussian–Lorentzian (GL) line shapes. The spin–orbit components of Cd 3d, Se 3d, and S 2p were fitted with constrained energy separations and peak-area ratios. Specifically, the spin–orbit splitting energies were constrained to approximately 6.8–7.0 eV for Cd 3d, ~ 0.9 eV for Se 3d, and ~ 1.4 eV for S 2p. The corresponding peak-area ratios were constrained according to the expected spin–orbit degeneracies, i.e., $\text{Cd } 3d_{5/2} / 3d_{3/2} \approx 3:2$, $\text{Se } 3d_{5/2} / 3d_{3/2} \approx 3:2$, and $\text{S } 2p_{3/2} / 2p_{1/2} \approx 2:1$. The same GL(30) line shape was employed for the individual spin–orbit components, and the FWHM values were constrained within narrow ranges of ~ 0.95 – 1.05 eV for Cd 3d, ~ 0.91 – 1.06 eV for Se 3d, and ~ 0.96 – 1.09 eV for S 2p. These constraints were applied consistently during the fitting process to minimize fitting subjectivity and improve reproducibility. The binding-energy positions were constrained within physically reasonable ranges based on the expected chemical states of Cd, Se, and S. The detailed fitting parameters and constraints are summarized in Table S3.

S7 Fabrication of Glycerol–Toluene Biphasic Liquid CQW Microsphere Lasers

The liquid microsphere lasers were fabricated in a glycerol–toluene biphasic system. 10 mL of glycerol was added to a clean glass Petri dish, and 1 mL of CQW–toluene dispersion was slowly dropped onto the glycerol surface. The mixture was gently stirred at 500 rpm for 10 minutes to promote uniform microsphere formation. Stirring rate, toluene-to-glycerol volume ratio, CQW doping concentration, and precursor addition sequence were optimized to obtain uniformly dispersed, monodisperse microspheres. The microspheres were left to stand for 30 minutes to stabilize the biphasic interface. The resulting liquid microsphere lasers were used for subsequent characterization and performance testing.

S8 Lasing Measurement and Setup

A micro-photoluminescence (μ -PL) system was employed to investigate the lasing behavior of CQW liquid microsphere lasers. A 355 nm Q-switched Nd:YAG laser, operating at a repetition rate of 20 Hz with a pulse width of 7 ns, was used as the excitation source. The 355 nm pump beam was focused onto the CQW liquid microspheres using a 10 mm focal-length focusing lens, resulting in a pump spot diameter of approximately 0.072 mm, corresponding to an illuminated area of

approximately 0.004 mm². The pump spot was positioned on the microsphere using a three-dimensional translation stage, and the same focusing configuration was maintained throughout the lasing measurements. The CQW liquid microspheres were mounted on a three-dimensional translation stage. The optical signals emitted from the functionalized microspheres were collected from the top using a 50× objective lens (NA = 0.42). The collected signals were then either directed to a camera for photoluminescence imaging or coupled into a spectrometer equipped with a silicon charge-coupled device (CCD) for spectral acquisition, with a spectral resolution of 0.043 nm.

S9 Mode analysis of CQWs-liquid microlaser

The lasing modes can be predicted from the following asymptotic equation [3-5]:

$$\lambda^{-1}(D, n_1, n_r, q, m) = \frac{1}{\pi D n_1} \left[m + \frac{1}{2} + 2^{-1/3} \alpha(q) \left(m + \frac{1}{2} \right)^{1/3} - \frac{L}{\sqrt{n_r^2 - 1}} + \frac{3}{10} 2^{-2/3} \alpha^2(q) \left(m + \frac{1}{2} \right)^{-1/3} - 2^{-1/3} L \left(n_r^2 - \frac{2}{3} L^2 \right) \frac{\alpha(q) \left(m + \frac{1}{2} \right)^{-2/3}}{(n_r^2 - 1)^{3/2}} \right]$$

in which λ denotes the resonant wavelength; R expresses the microcavity radius; n_1 represents the refractive index of the CQW liquid cavity ($n_1 = 1.59$ for CQWs in toluene), $n_r = n_1/n_2$, with n_2 as the refractive index of the surrounding medium ($n_2=1.47$ for glycerol); $L = 1/n_r$ for transverse magnetic (TM) modes and $L = n_r$ for transverse electric (TE) modes; $\alpha(q)$ expresses the roots of the Airy function, with q as the radial mode number.

S10 Numerical Approaches to Modeling electromagnetic-field distribution inside WGM Microcavity

The optical response of the WGM resonator was modeled using the finite element method implemented in the commercial software COMSOL Multiphysics. Numerical simulations were carried out in the Electromagnetic Waves, Frequency Domain (ewfd) interface of the Wave Optics Module, using two-dimensional axisymmetric models under Maxwell's boundary conditions. To ensure sufficient accuracy, the mesh size in the simulation was set to 80 nm. A perfectly matched layer (PML) together with scattering boundary conditions was applied at the outer boundaries of the model. The refractive indices of glycerol, toluene-CQWs, and air were set to 1.47, 1.59, and 1.00, respectively.

S11 Fluorescence microscopic characterization of liquid microspheres.

A fluorescence microscope (Olympus BX53) was used to characterize the morphology and dispersion behavior of the liquid microspheres. Specifically, the microspheres were deposited onto clean glass slides, and fluorescence images were acquired under optical excitation. The size of individual microspheres was then measured using ImageJ software, and the size distribution was statistically analyzed.

S12 Linear polarization test.

The polarization response of the laser emission was measured by inserting a polarizer between the sample and the spectrometer. The polarizer was rotated incrementally from 0° to 90° at intervals of 30°, and the PL intensity was recorded at each rotation angle. To quantify the degree of polarization (DoP) of the laser emission, the DoP value was calculated based on the measured PL intensity data, and the DoP is defined by the following formula:

$$DoP = \frac{I_{max} - I_{min}}{I_{max} + I_{min}}$$

where I_{max} is the maximum PL intensity obtained during the polarizer rotation, and I_{min} is the minimum PL intensity corresponding to the direction perpendicular to the polarization direction of the laser emission.

S13 Stability testing and calculation.

The long-term stability of the laser was evaluated under continuous pumping at an energy density of ~430 μJ/cm² (which is above the lasing threshold) for 800 minutes. To assess the stability, the PL intensity was normalized, and the spectral changes were systematically analyzed. Additionally, the Allan variance was calculated to quantitatively characterize the intensity fluctuations of the laser emission. The Allan variance is defined as:

$$\sigma^2(\tau) = \frac{1}{2(M-1)} \sum_{k=1}^{M-1} (\overline{y_{k+1}} - \overline{y_k})^2$$

where τ is the averaging (integration) time, M is the number of laser intensity measurements, and $\overline{y_k}$ represents the mean deviation of the laser output intensity over a time interval of duration τ .

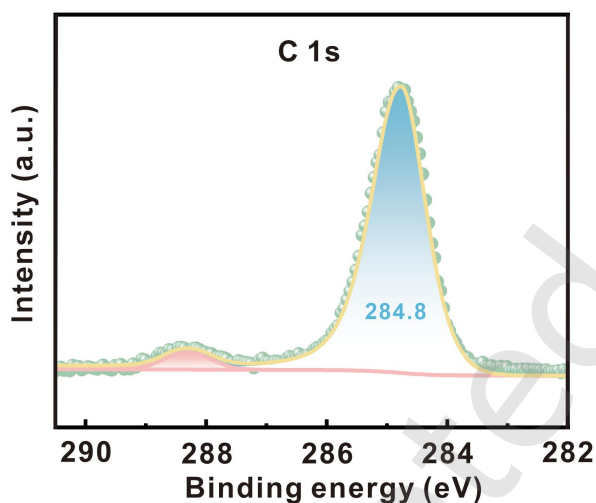


Figure S1. High-resolution C 1s XPS spectrum of green-emitting CdSe/CdSe_{1-x}S_x CQWs. The characteristic peaks at 284.8 eV correspond to C – C/C – H bonds from surface organic ligands.

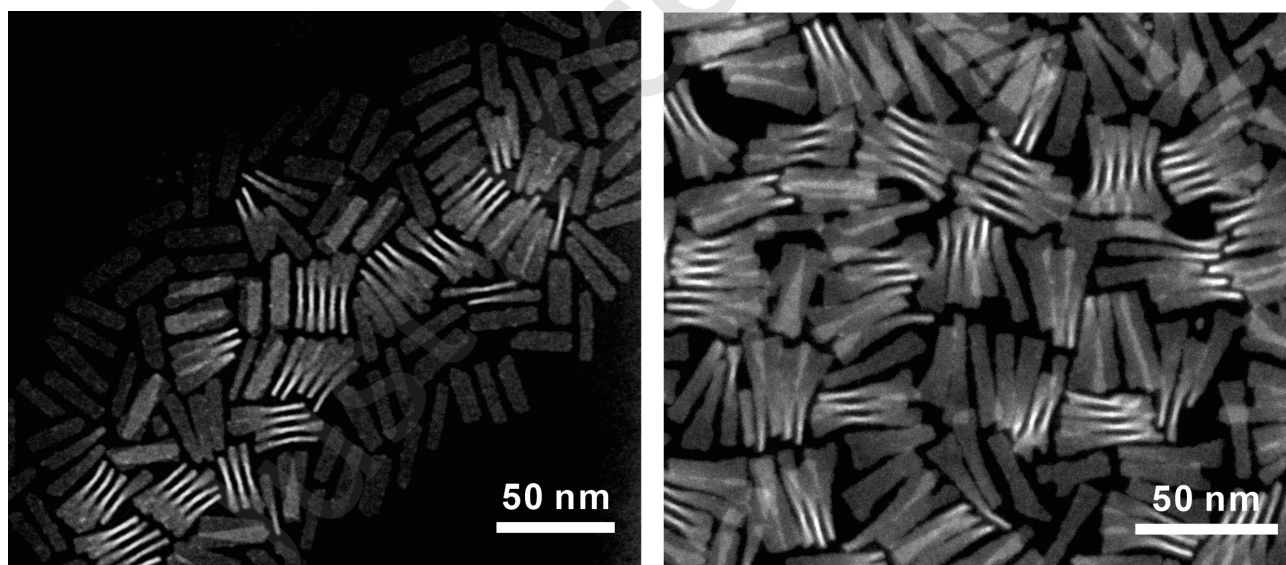


Figure S2. SEM image of green-emitting CdSe/CdSe_{1-x}S_x core/shell CQWs. The image reveals uniform one-dimensional nanostructures with good dispersibility and no obvious aggregation, confirming the successful synthesis of high-quality green-emitting CQWs.

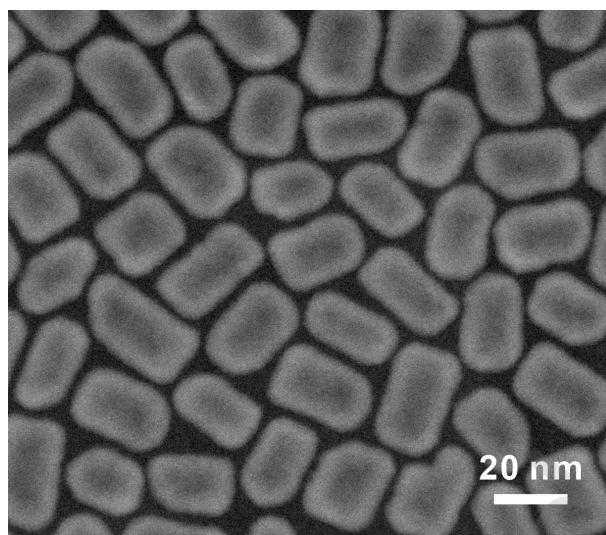


Figure S3. SEM image of red-emitting CdSe/Cd_{1-x}Zn_xS core/shell CQWs. The sample exhibits homogeneous morphology and narrow size distribution, demonstrating the high crystallinity and structural uniformity of the red-emitting CQWs.

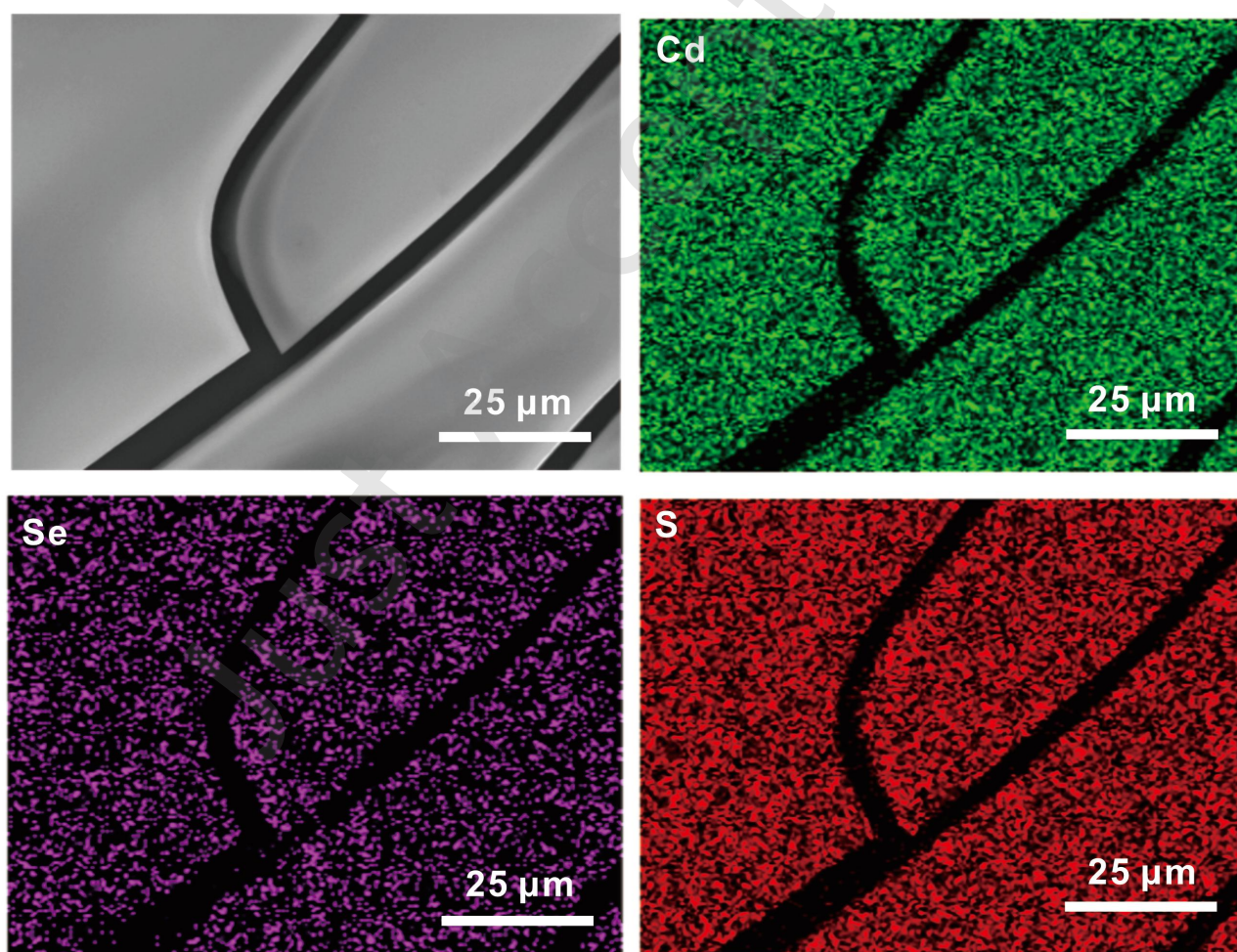


Figure S4. EDS elemental mapping and spectrum of green-emitting CdSe/CdSe_{1-x}S_x core-shell CQWs. The uniform spatial distribution of Cd, Se, and S elements verifies the formation of homogeneous core-shell structure and the effective incorporation of sulfur in the shell layer.

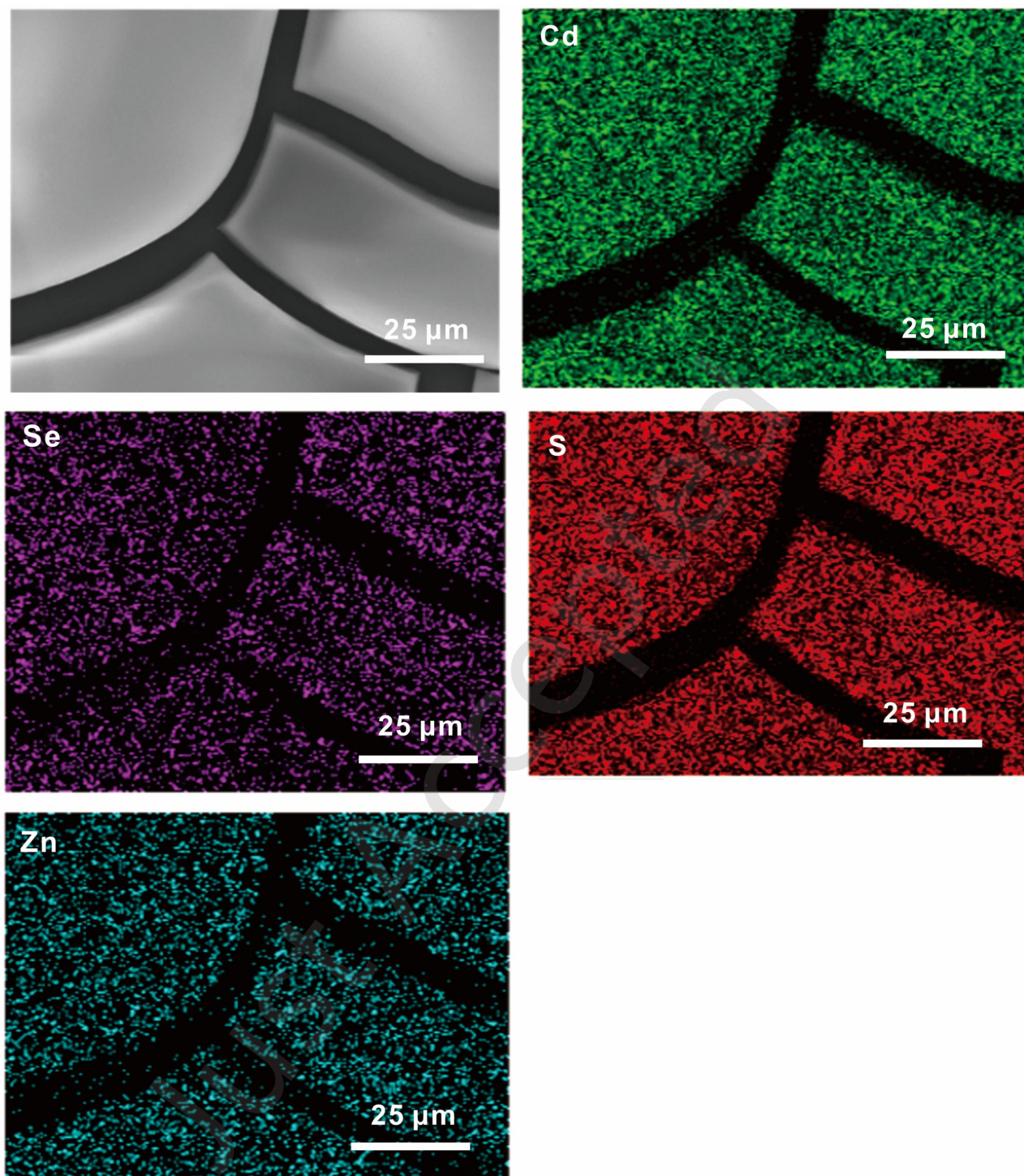


Figure S5. EDS elemental mapping and spectrum of red-emitting CdSe/Cd_{1-x}Zn_xS core/shell CQWs. The homogeneous distribution of Cd, Se, Zn, and S confirms the successful incorporation of Zn into the shell and the formation of uniform CdSe/Cd_{1-x}Zn_xS heterostructure.

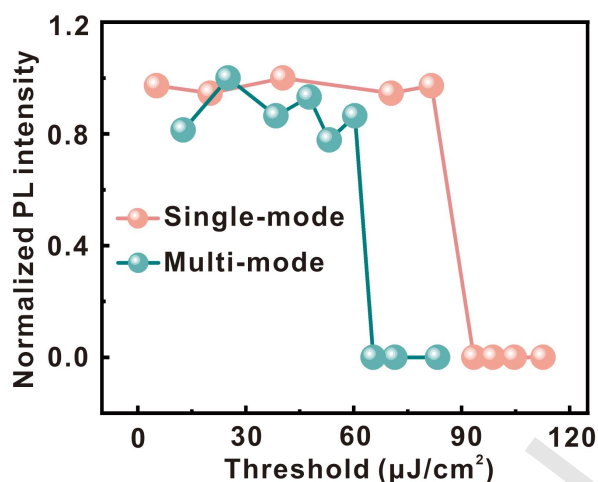


Figure S6. Lasing mode linewidth as a function of pump fluence for green-emitting liquid CQWs microsphere lasers. Single-mode lasing behavior is represented by the red curve, while multi-mode lasing is shown in green.

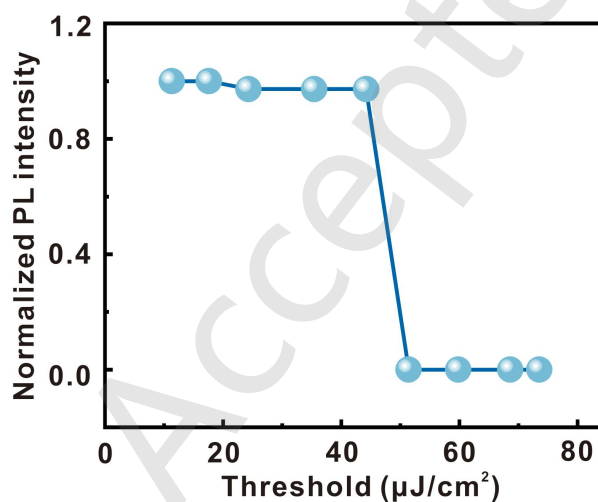


Figure S7. Single-mode lasing mode linewidth as a function of pump fluence for red-emitting liquid CQWs microsphere lasers. A clear lasing threshold is observed, and the linewidth narrows significantly above the threshold, demonstrating stable and low-threshold single-mode lasing performance from the red-emitting CQW-based liquid microsphere cavities.

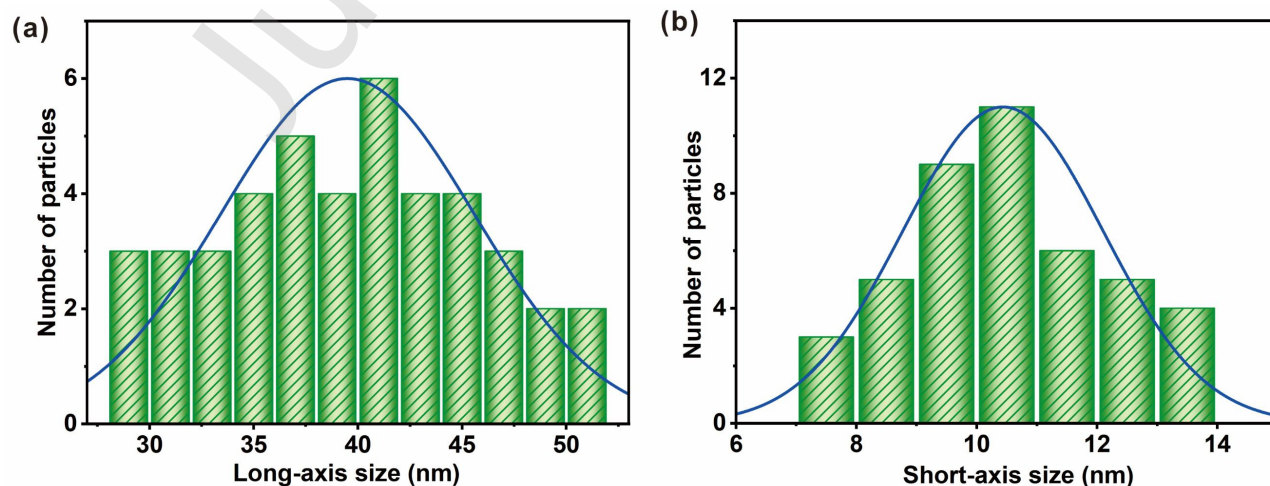


Figure S8. Histograms of the length and width distributions of the green-emitting CQWs.

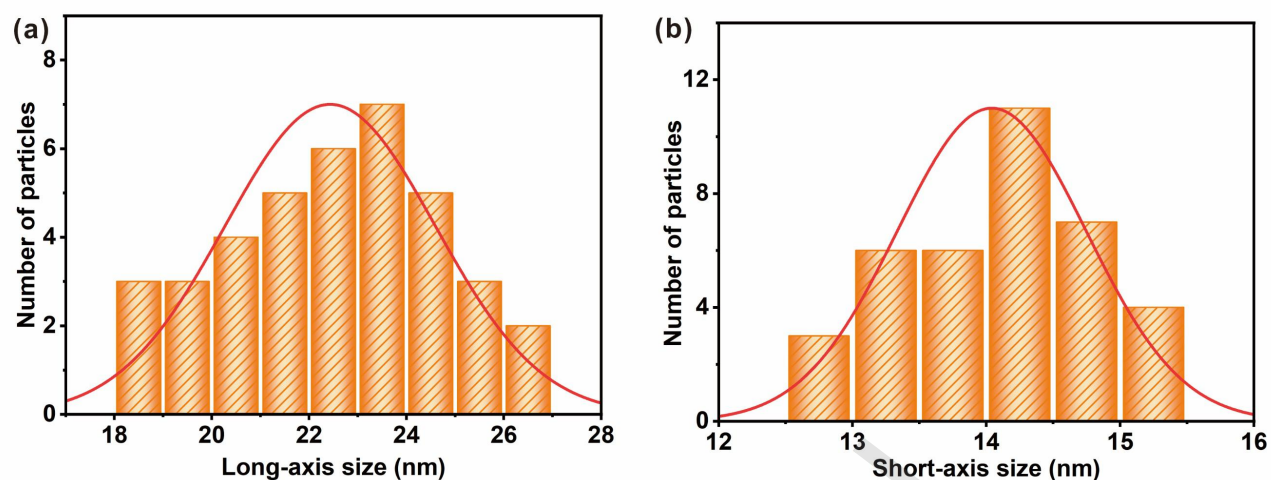


Figure S9. Histograms of the length and width distributions of the red-emitting CQWs.

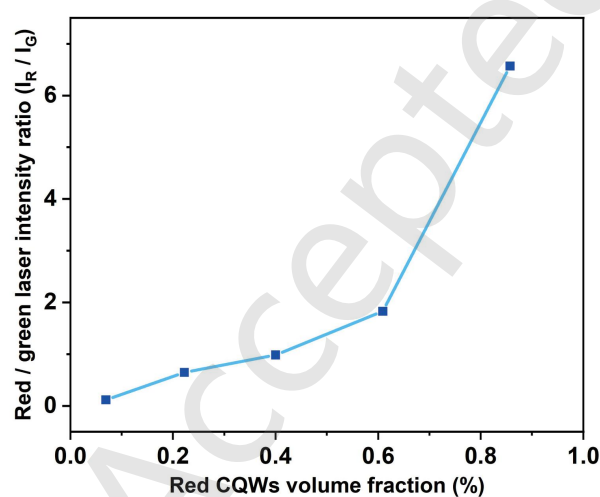


Figure S10. Red-to-green laser intensity ratio ($I_{\text{red}} / I_{\text{green}}$) as a function of the red CQW volume fraction at a constant total CQW concentration.

Table S1. TM/TE mode characteristic parameters of microsphere WGM lasers.

TM	Mode number	366	365	364	363	362	361
	Theoretical wavelength (nm)	516.19	517.58	518.98	520.38	521.79	523.21
	Experimental wavelength (nm)	516.40	517.70	518.83	520.22	521.62	523.01
TE	Mode number	366	365	364	363	362	361
	Theoretical wavelength (nm)	516.63	518.02	519.41	520.82	522.23	523.65
	Experimental wavelength (nm)	516.74	518.05	519.35	520.75	522.14	523.44

Table S2. Lasing features of liquid colloidal nanocrystal lasers.

Lasing type	Gain materials	Pumping laser type	Threshold	Q-factor	Ref.
F-P lasing	CdSe/CdS core-shell NPLs	Femtosecond	1.2 mJ/cm ²	≈1200	[1]
F-P lasing	CdSe/ZnSe/ZnSe _x S _{1-x} /ZnS core-shell	Nanosecond	50 mJ/cm ²	≈7500	[2]
F-P lasing	RQDs/OQDs/GQDs CdSe/ZnSe/CdS/ZnS type-(I+II) QDs	Nanosecond	44 mJ/cm ²	≈5300	[3]
F-P lasing	ZnSe-ZnS core-shell QDs	Nanosecond	1.14 mJ/cm ²	≈2100	[4]
WGM lasing	CdZnS/ZnS Ternary Alloy QDs	Nanosecond	210 μJ/cm ²	≈3000	[5]
WGM lasing	Dye-doped liquid crystals	Nanosecond	2.25 mJ/cm ²	≈1170	[6]
WGM lasing	CdSe/Cd _{1-x} Zn _x S C/S CQWs	Nanosecond	42.9 μJ/cm ²	≈32700	Our work

Table S3. XPS peak-fitting parameters and constraints for the high-resolution Cd 3d, Se 3d, and S 2p spectra.

Parameter	Cd 3d	Se 3d	S 2p
Spin-orbit splitting	~6.8 - 7 eV	~0.9 eV	~1.4 eV
Peak area ratio	Cd 3d _{5/2} : Cd 3d _{3/2} ≈ 3:2	Se 3d _{5/2} : Se 3d _{3/2} ≈ 3:2	S 2p _{3/2} : S 2p _{1/2} ≈ 2:1
Line shape	GL (30)	GL (30)	GL (30)
FWHM	~0.95-1.05 eV	~0.91-1.06 eV	~0.96-1.09 eV

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