Supporting Information

Low-threshold near-infrared lasing at room temperature

using low-toxicity Ag₂Se quantum dots

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Synthesis of Ag₂Se QDs:



Fig. S1 Schematic illustration of synthesis of Ag₂Se QDs.

Chemicals: Silver nitrate (AgNO₃, \geq 99.0%), oleylamine (OAM, 70%), selenium powder (Se, 99.99%), sodium borohydride (NaBH₄, \geq 96%) and 1-Dodecanethiol (DT, \geq 98.5%) were purchased from Sigma-Aldrich. Toluene, ethanol and tetracholoethylene were received from Sinopharm Chemical Reagent (Shanghai, China). All Chemicals were used as received without further purification.

Preparation of Ag-OAM: 34 mg (0.2 mmol) of AgNO₃ was dissolved in a mixture of 15 mL of OAM and 35 mL of toluene, and then heated to form an optically clear solution.

Preparation of NaHSe Solution: 39 mg (0.5 mmol) of Se powder and 38 mg (1 mmol) of NaBH₄ were added to 5 mL of pure water in a 25 mL three neck flask under nitrogen atmosphere. The mixture was stirred to form an optically clear solution to produce 0.1 mmol/mL of NaHSe, which was stored in nitrogen atmosphere.

*Synthesis of Ag*₂*Se QDs*: In a typical reaction, 35 mL of toluene containing 0.2 mmol of Ag-OAM was loaded into a 250 mL autoclave; then, 15 mL of DT and 35 mL of NaHSe solution (0.05 mmol) were added in order. The mixture solution was stirring for 5 min and then heated to 180 °C and aged for 1h. Upon cooling to room temperature, the products were washed by precipitating 2-3 times with ethanol. The final products were dispersed in tetracholoethylene for characterization, or in toluene for fabrication of thin film and microcavity.



Fig. S2 Scheme of transient absorption (TA) experiment.



Fig. S3 Sketch of amplified spontaneous emission (ASE) measurement.



Fig. S4 Experimental configuration for the variable stripe length (VSL) method.



Fig. S5 Sketch of laser performance characterization.



Fig. S6 Size histogram for Ag_2Se QDs taken from TEM image, and fitted by Gaussian distribution.



Fig. S7 Photoluminescence (PL) intensity of Ag₂Se QDs measured with increasing pump fluence (pump wavelength is 800 nm) and fitted with the function $\propto 1 - e^{-\sigma_{abs} j_p}$, where σ_{abs} is the QD absorption cross-section and j_p is the pump fluence (photons per cm²).^{1,2}



Fig. S8 The biexciton dynamics of the Ag₂Se QDs extracted by subtracting the low-fluence $(\langle N \rangle \ll 1)$ transient absorption trace from the "tail-normalized" trace measured for $\langle N \rangle$ slightly above unity.³ The fitted biexciton lifetime is 245 ps.



Fig. S9 Amplified spontaneous emission (ASE) intensity *versus* laser shots observed for an Ag_2Se QD film. The inset shows that the emission spectrum under a pump fluence of 424 μ J cm⁻² is dominated by ASE rather than spontaneous emission.



Fig. S10 Emission spectra of the coffee-ring microlaser in Fig. 4a recorded with pump fluence (238 μ J cm⁻²) above threshold.

Fabry-Perot cavity free spectral range (FSR) for the coffee-ring microlaser:

Modeling the cross section of the ring deposit as a standard Fabry-Perot cavity, the FSR can be calculated from:

$$FSR = \frac{\lambda^2}{2nL}$$

where λ , n, and L are the emission wavelength, the refractive index, and the cavity length, respectively. Utilizing a cavity length of 7.4 µm (as the full width at half-maximum (FWHM) of the deposit), we can estimate an FSR of 63.8 nm (assuming refractive index n = 2), obviously larger than the FWHM of the ASE peak (~44 nm), which is small enough to sustain single-mode emission.

Supporting Information References

- 1 F. Hu, H. Zhang, C. Sun, C. Yin, B. Lv, C. Zhang, W. W. Yu, X. Wang, X. Zhang and M. Xiao, *ACS Nano*, 2015, **9**, 12410-12416.
- 2 L. Zhang, H. Li, C. Liao, H. Yang, R. Xu, X. Jiang, M. Xiao, C. Lu, Y. Cui and J. Zhang, *J. Phys. Chem. C*, 2018, **122**, 25059-25066.
- 3 V. I. Klimov, A. A. Mikhailovsky, D. W. McBranch, C. A. Leatherdale and M. G. Bawendi, *Science*, 2000, **287**, 1011-1013.