Supplementary Information for

High Quality Two-Photon Pumped Whispering-Gallery-Mode Lasing from Ultrathin CdS Microflakes

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Tab.1 Comparison of TPP lasing dimension and threshold values reported semiconductor lasers with active CdS cavity.

Materials	Thickness	λ (nm)	Threshold (mJ/cm^2)	Excitation sources	Ref.
CdSe/CdS NP film	------	534	4.4-8.3	800 nm, 1 kHz,120 fs	1
CdSe/CdS NR film	Thickness: 500 nm	550-600	1.5 -3.4	800 nm, 1 kHz, 150 fs	2
CdS MW	Dimeter:1 μm Length:28 μm	523	3.3	800nm, 1 kHz, 80 fs	3
CdS NW	Dimeter: ~100 nm	513	3.15	780nm, 1 kHz, 1.6 ps	4

*Abbreviations: NP, nanoplatelet; NR, nanorod; MW, mircowire; NW, nanowire.[1-4]

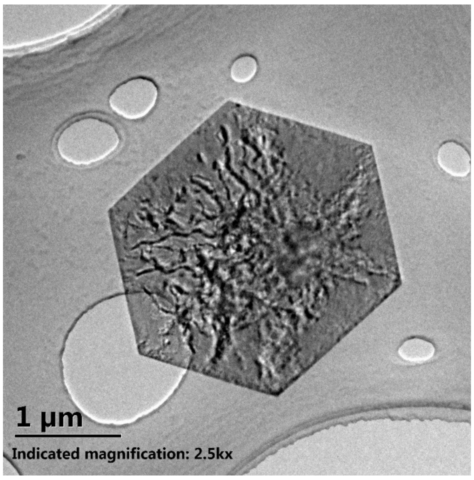

Fig. S1 Low-magnification TEM image of a hexagonal CdS microflake.

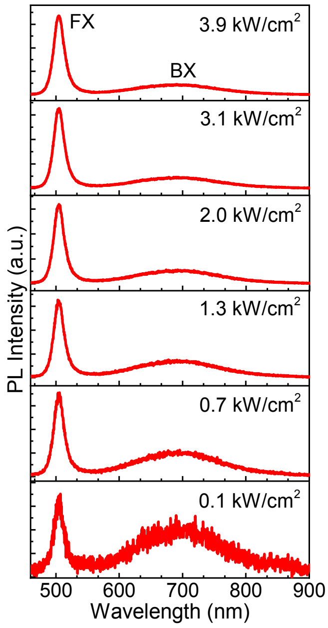

Fig. S2 PL spectroscopy of a CdS microflake at power density of 0.1, 0.7, 1.3, 2.0, 3.1 and 3.9 kW/cm^2, respectively. Excitation source is a CW laser with a wavelength of 405 nm.

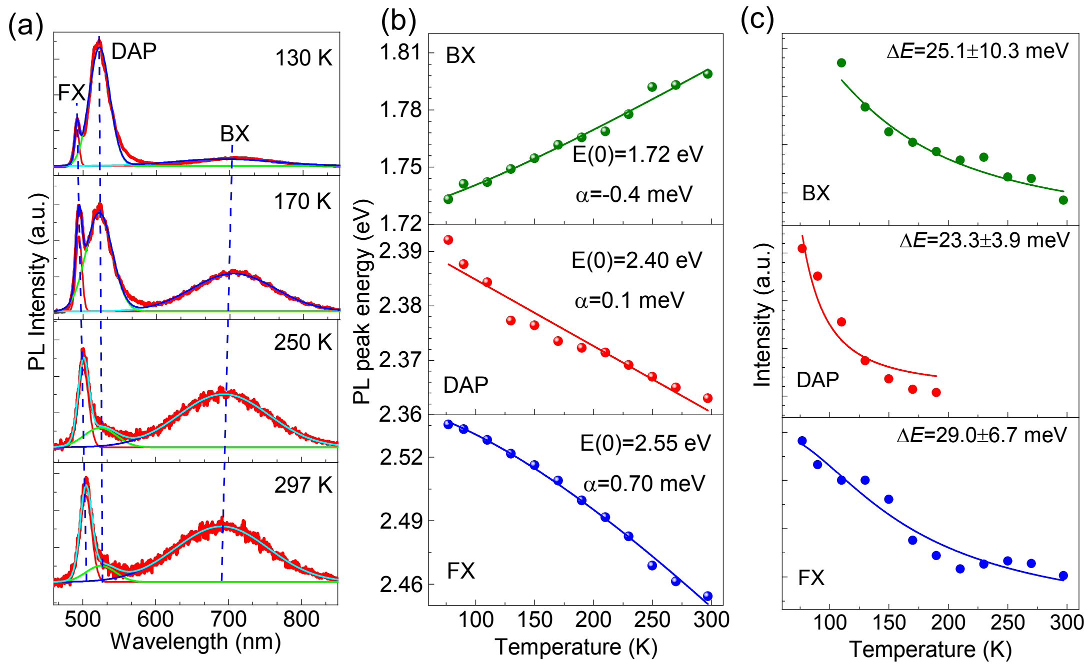

Fig. S3 (a) PL spectroscopy of the CdS microflake at 297, 250, 170, and 130 K. (b) Energy shifts of FX, DAP and BX emission with temperature. Scatters: data points; solid lines: fitting curves using function of $E(T) = E(0) - \alpha T^2/(T+\delta)$, where $E(0)$ is the bandgap at 0 K, α represents the liner shift of $E(T)$ at high temperature, and δ stands for the quadratic variation of $E(T)$ at low temperature. (c) PL intensity of FX, DAP and BX emission as a function of temperature. Scatters: data points; solid lines: fitting curve using a function of $I(T) = I_0/(1+\mathrm{C}e^{-\Delta E/k_\mathrm{B}T})$, where I_0, C and k_B represent the PL intensity at 0 K, a constant and the Boltzmann constant, respectively. The activate energies ΔE of FX, DAP and BX are 29.0±6.7, 23.3±3.9 and 25.1±10.3 meV, respectively.

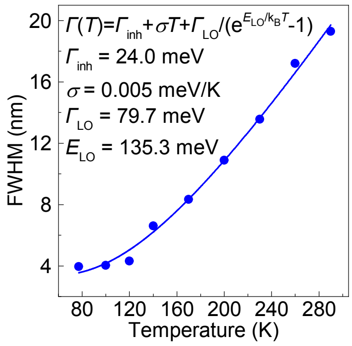

Fig. S4 Temperature-dependence FWHM of a typical CdS microflake. Scatters: data points; solid and dashed lines: fitting curve using Boson model $\Gamma(T) = \Gamma_{\mathrm{inh}}+\sigma T+\Gamma_{\mathrm{LO}}/(e^{E_{\mathrm{LO}}/k_{\mathrm{B}}T}-1)$, where Γ_{inh} the inhomogeneous broadening factor, σ is exciton-longitudinal acoustic phonon coupling coefficient, Γ_{LO} is exciton-longitudinal optical (LO) phonon coupling coefficient, and E_{LO} is phonon energy.

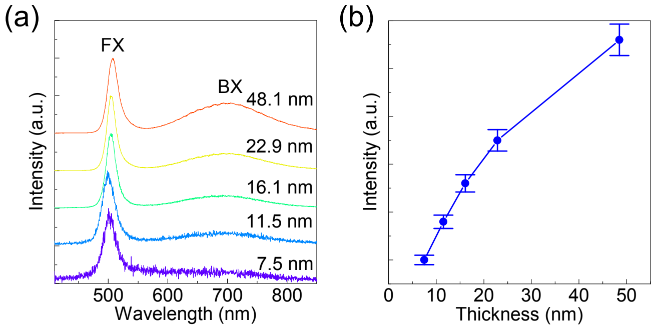

Fig. S5 (a) Normalized PL spectra of CdS microflakes with thickness from 7.5 to 48.1 nm at a fixed average power density of 1.0 MW/cm^3 under 405 nm laser. (b) Plot of integrated PL intensity of BX emission of the CdS microflakes in (a).

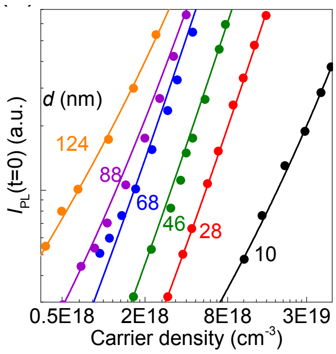

Fig. S6 Photon-generated carrier density dependence of the initial time PL intensity (I_{PL}(t=0)) of CdS with thickness of 10, 28, 46, 68, 88, and 124 nm, respectively, following excitation at 400 nm (80 MHz, 140 fs).

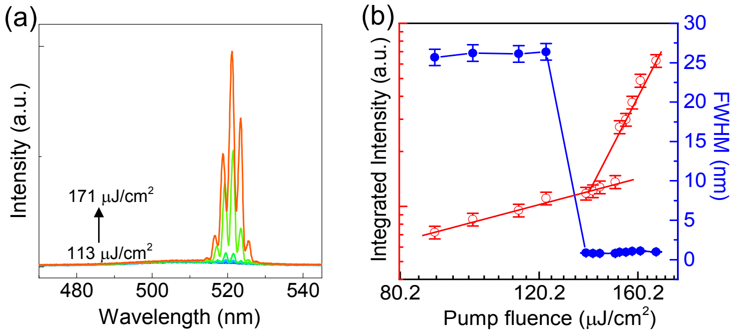

Fig. S7 (a) The evolution from spontaneous emission to lasing in a typical CdS microflake using 400 nm pulse laser as excitation source. The pumping fluence increased from 113 to 171 μJ/cm^2. (b) Power-dependence of the integrated PL intensity and FWHM of the microflake as a function of power fluence, which gives a threshold of ~137.8 μJ/cm^2.

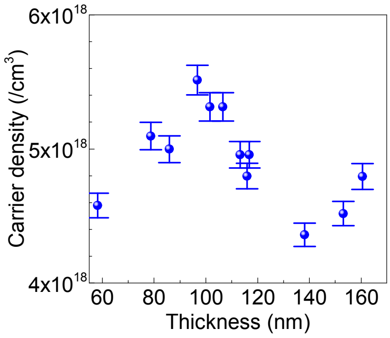

Fig. S8 The calculated photo-generated carrier density under pulsed 800 nm laser at lasing threshold for CdS microflakes with different thicknesses (blue scatters).

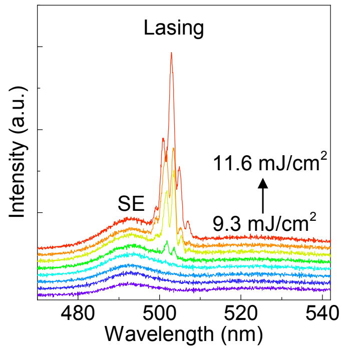

Fig. S9 Emission spectra of microflake (thickness: 46.0±0.3 nm) with a threshold of 10.8 mJ/cm^2 at pump fluences from 9.3 to 11.6 mJ/cm^2.

Supplementary Note 1: Estimation of average power density, carrier density and defect density

The average power density P_{ave} in CdS microflakes can be expressed as:

$$P_{ave} = P(1-R)(1-exp(-\alpha d))/(\pi r^2 d))$$

where P is excitation power, $r = 1$ μm is laser spot, $R = 0.2$ is the reflection coefficient of CdS, d is the thickness of the microflakes, and $\alpha \sim 1.2\times10^5$ cm^{-1} is the absorption coefficient of CdS at 405 nm. For a fixed average power density of 1.0 MW/cm^3, the P are 30.3, 46.5, 66.7, 92.7, and 196.9 μW for CdS microflakes with thicknesses of 7.5, 11.5, 16.5, 22.9, and 48.1 nm, respectively.

For photon-generated carrier density excitation by pulsed laser,[5]

$$N = \frac{E_{ave}\cdot(1-R)}{2\cdot\hbar\cdot\omega\cdot d}\cdot(1-exp(-\alpha P_{peak}\cdot d))$$

where $R = 0.2$ is the reflection coefficient of CdS, d is the thickness of the microflakes, $\hbar\omega$ is the incident photon energy, E_{ave} is average pump energy (mJ/cm^2), $P_{\text{peak}} = P_{\text{ave}} f_{\text{pump}}/\Delta t$ is peak pump power (GW/cm^2), P_{ave} is average pump power, f_{pump} is repetition rate, Δt is pulse duration, and α is absorption coefficient (2.7 cm/GW for two photon absorption at 800 nm).[6] The photon-generated carrier density is an order of 10^{18} cm^{-3} at threshold under pulsed 800 nm laser (1 kHz, 80 fs) (Fig. S8).

The initial time PL intensity ($I_{\text{PL}}(t = 0)$) could be expressed as:[7]

$$I_{PL}(t=0) = k_1 n(0) + \frac{k_2 n^2(0)}{n_t} + n(0)(k_1 - k_2)e^{-n(0)/n_t}$$

where $n(0)$ is the photo-generated carrier density, n_{t} is the defect density, k_1 and k_2 are the exciton and defect recombination coefficients. Fitting the $I_{\text{PL}}(t = 0)$ with the equation, as shown in Fig. S6, the defect density of CdS microflakes with thickness of 10, 28, 46, 68, 88, 124 nm are estimated to be around 1.0×10^{15}, 1.2×10^{15}, 2.2×10^{15}, 3.9×10^{15}, 4.1×10^{15} and 4.2×10^{15} cm^{-3}, respectively.

References

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