Supporting Information for

Unveiling Lasing Mechanism in $CsPbBr_3$ Microspheres Cavities

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Contents of the Supporting Information:

I. The setup of chemical vapor deposition method

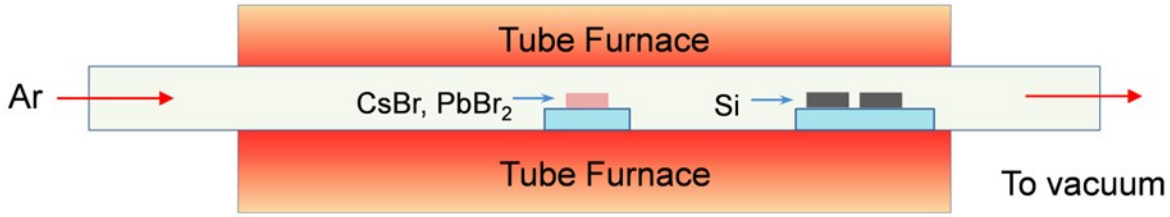

Figure S1 | Schematic of the chemical vapor deposition method setup

$PbBr_2$ and CsBr powders (with 1:2 stoichiometry) were placed in the heating center of a quartz tube reactor. The silicon (Si) substrate was positioned at a distance of 12 cm away from the powder sources in the downstream. Ar as the carrier gas is flowing from the upstream to the downstream. At a sample temperature of 575 ℃, we observed the reliable growth of $CsPbBr_3$ perovskite micro-crystals with a special shape of microspheres.

II. SEM images of sphere samples with different titled angle

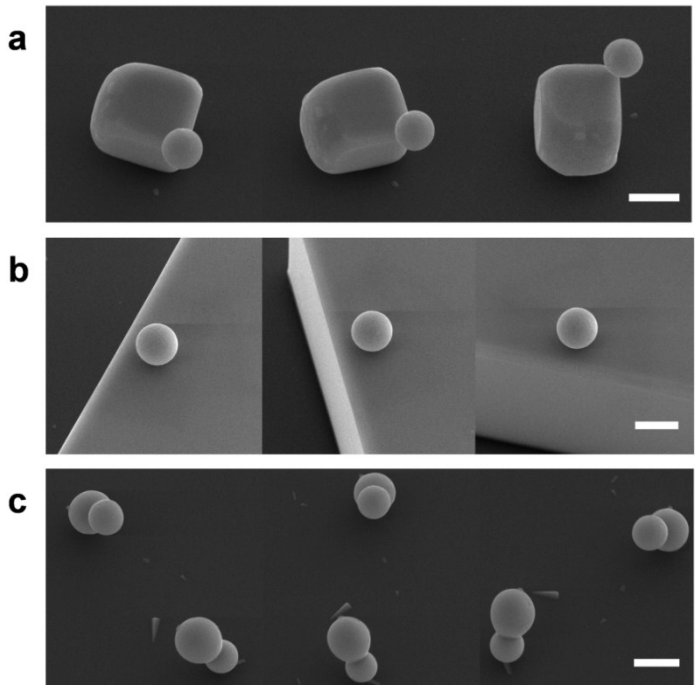

Figure S2 | SEM images of three sphere samples with different titled angle. The error bar is 1 μm.

III. Estimation of exciton binding energy

The exciton binding energy can be fitted using equations, [1-2] $I(T) = R\left[1 - exp\left(-\frac{E_b}{k_B T}\right)\right] + c$, where *I(T)* is the integrated PL intensity at a specific temperature *T*. Therefore, temperature-dependent steady-state PL spectroscopy is conducted on individual $CsPbBr_3$ microsphere in a backscattering configuration with an excitation laser of 405 nm. The temperature is varied from 77 K to 300 K. The integrated PL intensity versus 1/*T* of four samples are plotted in **Figure S3** (solid black dots), showing the exciton binding energy from 65 ~ 86 meV. These values are much higher than the thermal disturbance at room temperature (*KT*~ 26 meV).

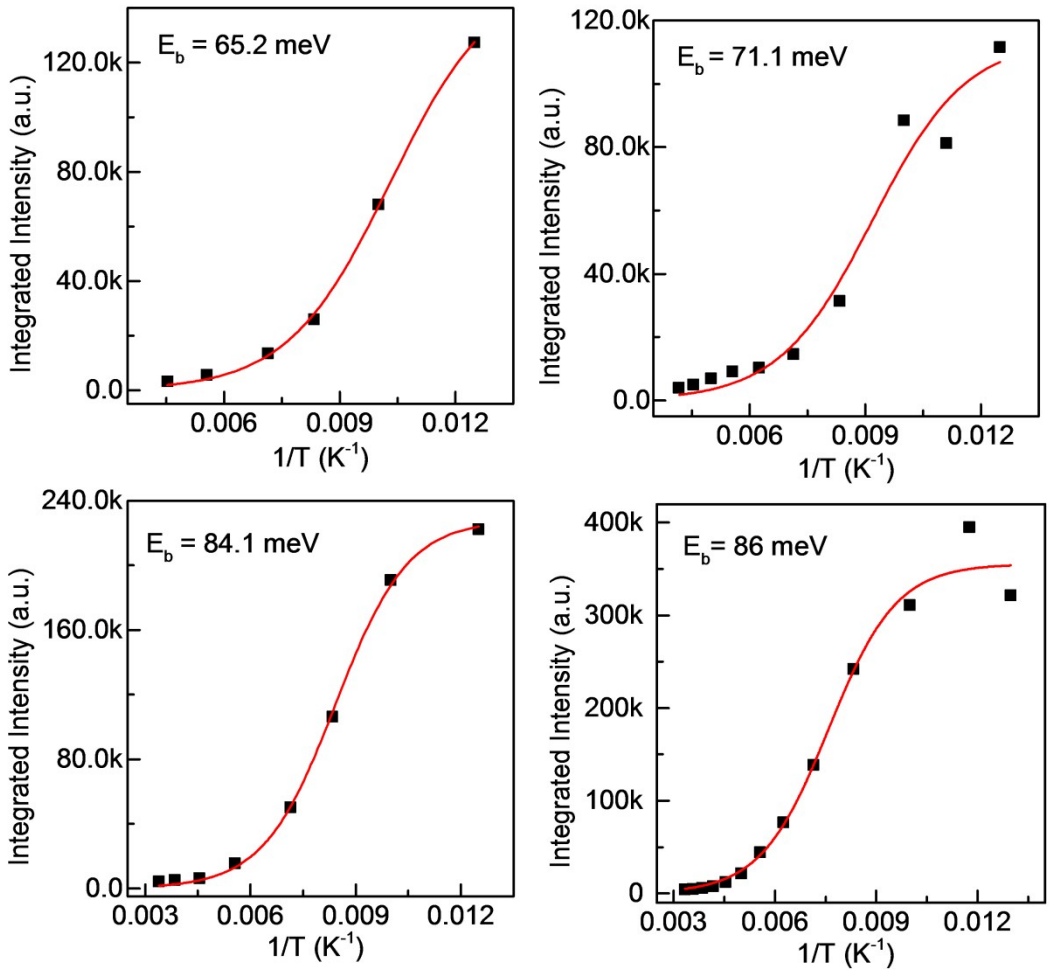

Figure S3 | The integrated PL intensity of four samples as a function of *1/T*. The experimental data are well fitted.

IV. The power dependent PL

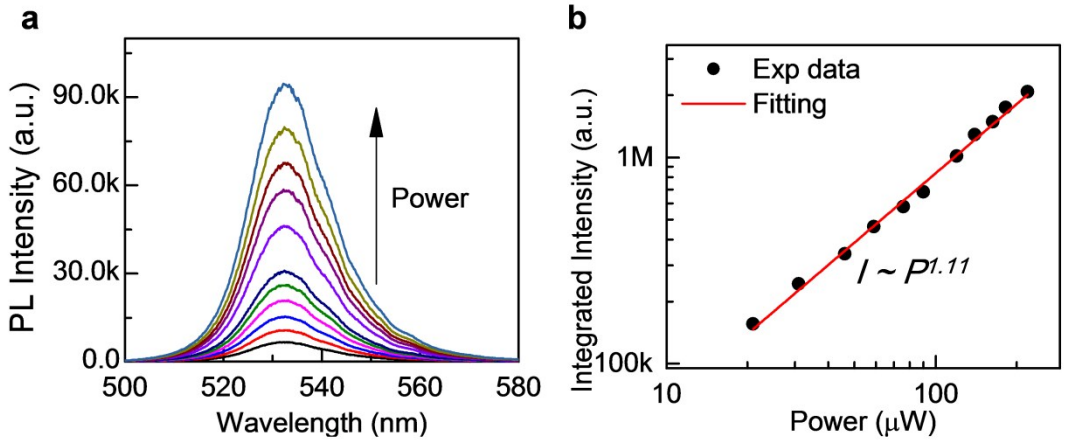

Figure S4 | (**a**) PL spectroscopy of $CsPbBr_3$ microsphere at room temperature with different laser power from 21 μW to 220 μW. (**b**) PL intensity as a function laser power in a log-log plot. The red line is a power-law fit $I=P^k$. Linear fitting of these plots gives a slope of 1.11 ± 0.03, confirming exciton recombination process in these microspheres.

V. Temperature-dependent Raman spectra

To confirm the exciton-phonon coupling in sphere microcavities, Raman measurement is executed using 633 nm laser as the excitation source (**Figure S**5). As the $CsPbBr_3$ microsphere is heated from 103 K to 303 K, the splitting peaks near 70 cm^{-1} of the Raman spectra are undistinguishable by degrees.

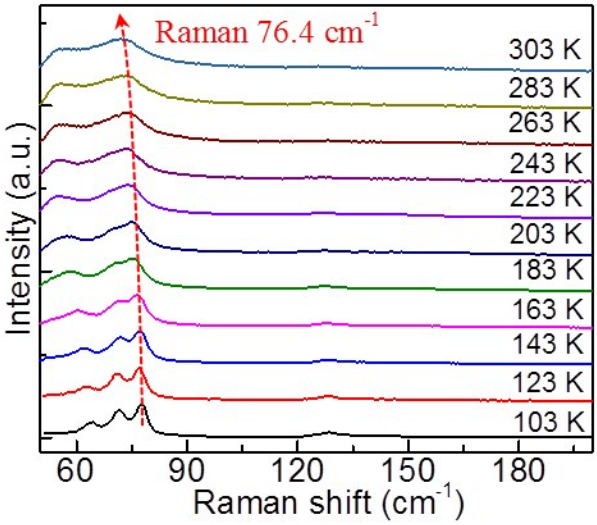

Figure S5 | Raman spectra of a typical $CsPbBr_3$ sphere at different temperatures from 103 to 303 K, showing the Raman shift as the temperature changes.

VI. The anharmonic model

In the anharmonic model, the variation of phonon frequency $\omega(T)$ and width $\Gamma(T)$ can be described by an anharmonic expression:

$$\omega(T) = \omega_0 + \Delta(T) \quad \text{(S1)}$$

$$\Delta(T) = A\left\{1 + \frac{2}{e^x - 1}\right\} + B\left\{1 + \frac{3}{e^y - 1} + \frac{3}{(e^y - 1)^2}\right\} \quad \text{(S2)}$$

for temperature dependence of wave number at 76 cm^{-1}, and

$$\Gamma(T) = C\left\{1 + \frac{2}{e^x - 1}\right\} + D\left\{1 + \frac{3}{e^y - 1} + \frac{3}{(e^y - 1)^2}\right\} \quad \text{(S3)}$$

for temperature dependence of line-width at 76 cm^{-1}, where

$$x = \hbar\omega_0 / 2K_BT \quad (6)$$

$$y = \hbar\omega_0 / 3K_BT \quad (7)$$

ω_0 is the characteristic frequency of the mode; A, B, C and D are constants.

VII. Time-resolved PL spectroscopy and fitting

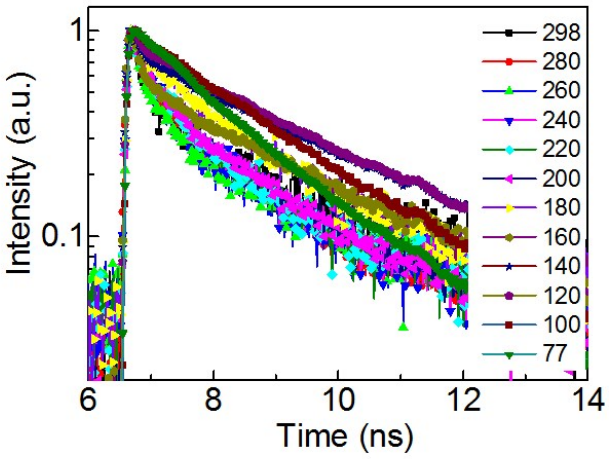

Figure S6 TRPL decay curves of $CsPbBr_3$ spheres with different temperatures from 77 K to 298 K monitored around 530 nm with excitation at 400 nm (76 MHz, 120 fs).

The PL decay curve in **Figure S**6 can be well fitted by multi-exponential decay equation as follows:

$$I(t) = I_0 + A_1 \cdot e^{-\tau_1/t} + A_2 \cdot e^{-\tau_2/t} \quad \text{(S4)}$$

$$T = \left. A_1 * \tau_1^2 + A_2 * \tau_2^2 \right/ A_1 * \tau_1 + A_2 * \tau_2 \quad \text{(S5)}$$

where I, T, τ_i, A_i are the integrated PL intensity, average lifetime, lifetime and pre-exponential factors of $CsPbBr_3$ spheres at different temperature. The fitting results are shown in **Table S1**.

Table S1 The fitting results by multi-exponential decay equation

	298K	280K	260K	240K	220K	200K	180K	160K	140K	120K	100K	77K
τ_1(ns)	2.26	1.63	1.61	1.52	1.62	1.95	1.98	2.69	2.98	2.80	2.32	2.78
τ_2(ns)	0.23	0.18	0.21	0.23	0.25	0.23	0.31	0.14	0.15	0.12	1.12	1.32
A_1 (ns)	0.19	0.26	0.19	0.28	0.25	0.26	0.19	0.27	0.37	0.24	0.10	0.13
A_2 (ns)	0.39	0.41	0.40	0.34	0.32	0.37	0.34	0.44	0.26	0.42	0.41	0.41

VIII. The setup of optical measurement system

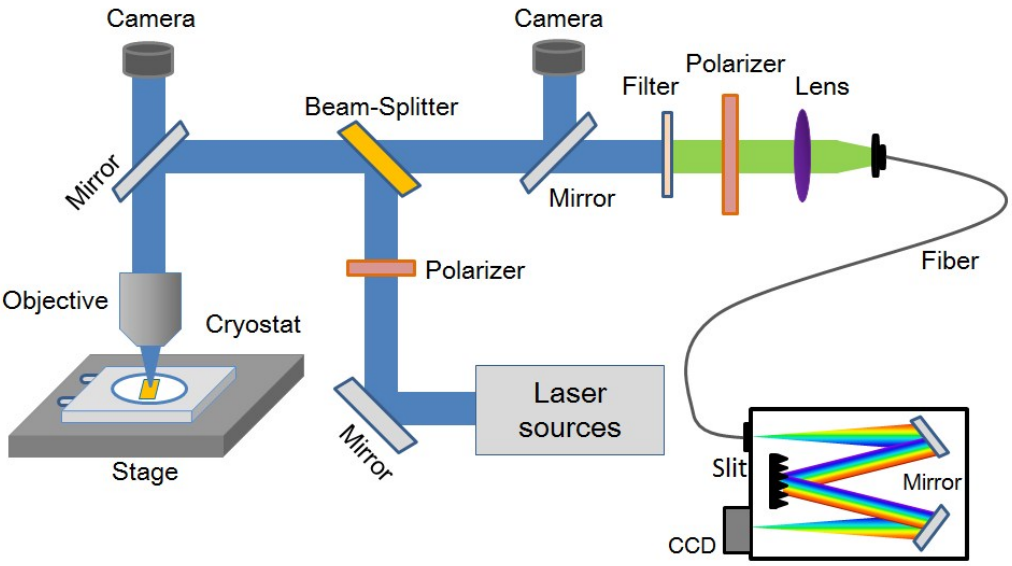

Figure S7 The setup of optical measurement system. For steady-state PL spectroscopy, a CW laser (wavelength: 405 nm) is focused onto an individual $CsPbBr_3$ sphere using an Olympus BX51 microscope equipped with a 100× objective (NA = 0.95). PL emission signal was collected by the same microscope objective in a backscattering configuration and analyzed by Princeton Instrument spectrometer (PI Acton, Spectra Pro 2500i) equipped with a TE-cooled charge coupled detector camera (PIXIS-400B). The PL image is recorded by a cool-snap color camera equipped on Olympus BX51 microscope. The PL lifetime measurements were conducted by a time-corrected single photon counting technique (TCSPC) with ultimate temporal resolution of ~40 ps. The excitation laser source is a frequency-doubled mode-locked Ti-sapphire oscillator laser (800 nm, repetition rate 76 MHz, pulse length 120 fs). For lasing measurements, the laser source is generated by frequency doubled from a Coherent Astrella regenerative amplifier (80 fs, 1 kHz, 800 nm) that was seeded by a Coherent Vitara-s oscillator (35 fs, 80 MHz).

References:

(1) Chen, Z.; Yu, C.; Kai, S.; Wang, J. J.; Pfenninger, W.; Vockic, N.; Midgley, J.; Kenney, J. T., Photoluminescence study of polycrystalline CsSnI 3 thin films: Determination of exciton binding energy. *J. Lumin.* **2012,** *132*, 345-349.

(2) Zhang, Q.; Ha, S. T.; Liu, X.; Sum, T. C.; Xiong, Q., Room-Temperature Near-Infrared High-Q Perovskite Whispering-Gallery Planar Nanolasers. *Nano Lett.* **2014,** *14*, 5995-6001.