Electronic Supplementary Material (ESI) for Nanoscale.

Supporting information

Investigation of Random Lasing from All-Inorganic Halide Perovskite Quantum Dots Prepared at Ambient Condition

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Temperature dependent PL of $CsPbBr_3$ QDs film

Figure S1(a) shows the normalized two-dimensional (2D) map of $CsPbBr_3$ QDs film from 77 K to 300 K. Unlike variation of PL from typical semiconductor showing red-shift with temperature because of the electron-phonon interaction,[1-2] the PL peak of $CsPbBr_3$ QDs film reveals blue-shift owing to the thermal expansion of lattice.[3] Figure S1(b) shows the variation of optical band gap (E_g) of $CsPbBr_3$ QDs film as temperature increases from 78K to 300K, which can be described by [4-5]

$$E_g(T) = E_0 + A_{TE}T + A_{EP}[\frac{2}{exp\left(\frac{\hbar\omega}{K_BT}\right)-1} + 1], \qquad \text{(S1)}$$

where E_0 is the band gap at 0 K, A_{TE} and A_{EP} are the weight of the thermal expansion and electron-phonon interaction, respectively. $\hbar\omega$ is the average optical phonon energy, K_B is Boltzmann constant. By the fitting of Eq. (S1) (red solid curve in Fig. S1(b)), four parameters, E_0 = 2.364 eV, A_{TE} = 0.263meV/K, A_{EP} = - 66 meV, $\hbar\omega$= 59 meV, can be obtained. The red solid curve can be decomposed by the two curve, includes green dash line and blue dash-dot curve to illustrate the thermal expansion and electron-phonon interaction, respectively. Below 140 K, the linear-increase of PL peak was dominated by A_{TE} and A_{EP} was negligible due to the unsubstantial populated optical phonon modes. For the higher temperature, the optical phonon (blue dash line in Fig. S1(b)) modes are appreciably populated, leading to a reduction of optical band gap.

Figure S1(c) shows the temperature-dependent FWHM of $CsPbBr_3$ QDs film which can be described by equation[4, 6]

$$\Gamma(T) = \Gamma_{inh} + \sigma\mathrm{T} + \frac{\Gamma_{LO}}{\exp\left(\frac{E_{LO}}{K_BT}\right)-1}, \qquad \text{(S2)}$$

where Γ_{inh} is the inhomogeneous broadening and the other two terms contribute to the

homogeneous broadening. Here, σ, Γ_{LO}, K_B and E_{LO} are the exciton–acoustic phonon coupling coefficient, exciton–longitudinal optical (LO) phonon coupling coefficient, Boltzmann constant. and the LO phonon energy, respectively. From the best fitting by the Eq. (S2) (red solid curve in Fig. S1(c)), we can also obtain the parameters Γ_{inh}= 61.3 meV, σ= 71.6 μeV/K, Γ_{LO} = 90.2 meV, E_{LO} =58.9 meV. In general, the electron - LO phonon interaction (Γ_{LO}) is much larger than the interaction between electrons - acoustic phonons (σ), which can almost be negligible for the variation of FWHM.

In Fig. S1(d), the peak intensity decreases with the increase of temperature, which indicates the thermal quenching of PL of $CsPbBr_3$ QDs film. In general, the temperature dependence of the PL intensity can be accounted for the Arrhenius equation[7]

$$I(T) = \frac{I_0}{1+Aexp(-E_B/K_BT)}, \quad \text{(S3)}$$

where I_0 is peak intensity at 0 K, K_B is Boltzmann constant and E_B is binding energy. By the well-fitting of Eq. (S3), the binding energy (E_B) of $CsPbBr_3$ QDs is around 49 meV, which is very close to previously report[1] around 50 meV.

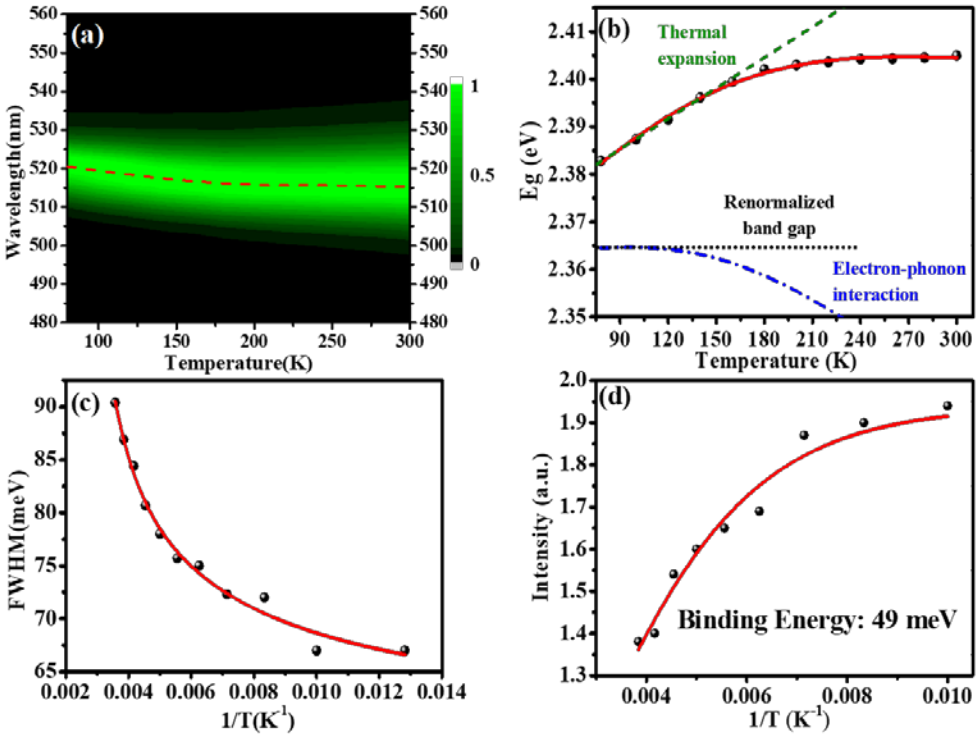

Figure S1. Temperature-dependent PL measurement of $CsPbBr_3$ QDs film with (a) two-dimensional (2D) map of the normalized PL, (b) band gap, (c) FWHM, and (d) PL peak intensity.

Gain measurement of $CsPbBr_3$ QDs film

The optical gain is an important parameter for assessing the possibility of realizing random lasing. The optical gain coefficients of the $CsPbBr_3$ QD film were measured via the variable stripe length (VSL) method. In Fig. S2(a), the sample was excited by stripe pump beam which was focused by a cylindrical lens. The length of stripe was controlled by a moving blade. The variation of the integrated ASE intensity as a function of stripe length is shown in Fig. S2(b) that can be well fitted by the following equation[8-9]

$$I(L) = (A/g)[\exp(gL)-1], \qquad \text{(S4)}$$

where I, A, g, and L are the detected amplified spontaneous emission (ASE) intensity, the cross-sectional excited area mapped by the penetration depth, gain coefficient, and excitation stripe length of $CsPbBr_3$ QDs, respectively. By the fitting of Eq. (S4), the gain coefficient of S-I and S-II are around 64 cm^{-1} and 73 cm^{-1}.

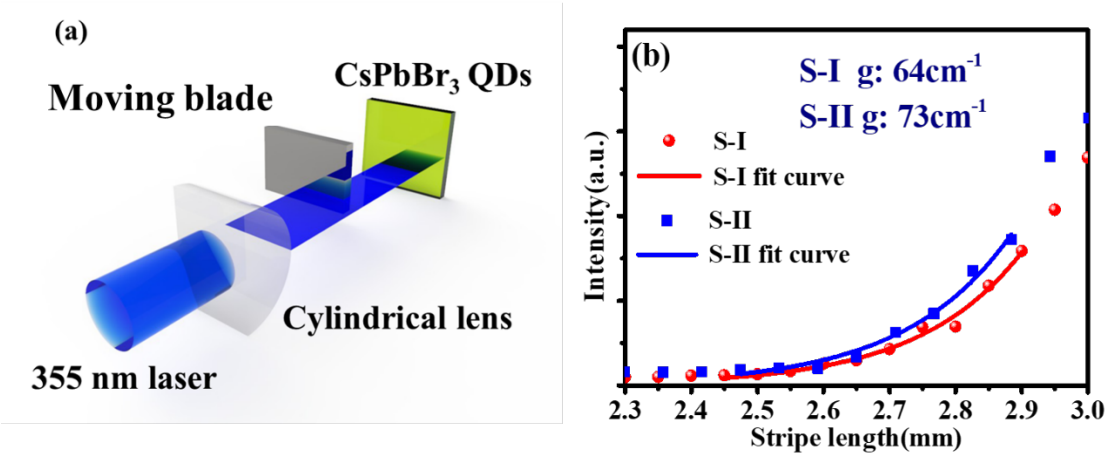

Figure S2. Gain measurement from $CsPbBr_3$ QDs by means of the variable stripe method with (a) schematic setup, and (b) ASE peak intensity as a function of stripe length from S-I (red circles) and S-II (blue circles). Red and blue solid line show the fitting curves by the Eq (S4).

Random lasing emission spectrum at different angle

Figure S3(a)-(c) show the random lasing spectrum with the angle θ relative to the edge direction of the sample. It is obviously to see a number of spikes on top of emission spectrum at different angle. The setup in Fig. S3(c) illustrates the pump excitation from 355 nm Q-switched laser and the measured direction relative to the edge direction of sample.

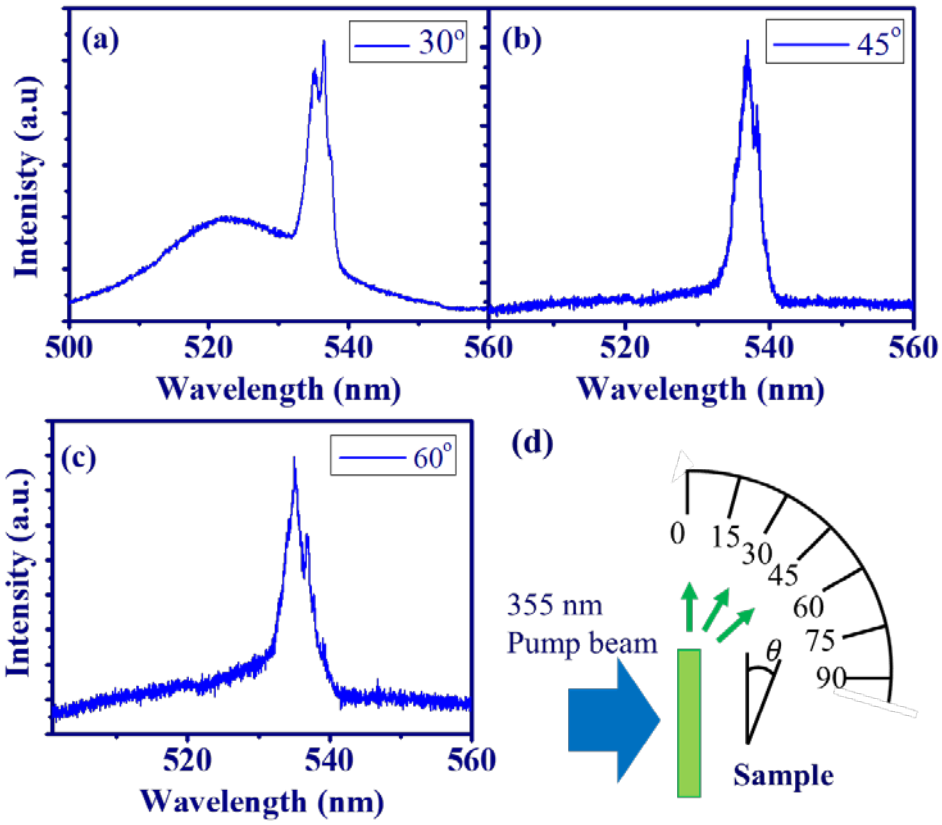

Figure S3 Random lasing spectrum with angle (a) θ=30°, (b) θ=45°, (c) θ=60° relative to the edge of the perovskite. (d) Schematic illustration to describe the setup for the random laser generation and the measured direction relative to the edge direction of sample.

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