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# Supporting Information for: "Femtosecond Laser Direct Writing of Perovskite Patterns with Whispering Gallery Mode Lasing"

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## CONTENTS

## S1. Details on Thermal Co-evaporation of $FAPbI_3$ Perovskite Films

Two source materials, *i.e.,* formamidinium iodide ($CH_3NH_3I$, FAI) and lead iodide ($PbI_2$), were placed in an organic crucible and inorganic crucible, respectively. The deposition rate of each crucible was monitored using a quartz crystal microbalance (QCM). The substrates were placed in a substrate holder above the crucibles with the clear quartz glass side facing down towards two sources. The rate of $PbI_2$ was controlled around 1 ± 0.1 Å $s^{-1}$ at ~250$^oC$. The substrate holder was kept at 130$^oC$ and rotated with a speed of 300 rpm, ensuring uniform deposition. We have carefully optimized the substrate temperature, and found that with the increase of temperature from 25$^oC$ to 130$^oC$, lead iodide can react nicely with FAI and form black phase perovskite crystals. However, with the further increase of substrate temperature, the formed perovskite thin film will decomposite back to lead iodide and FAI to some extent under vacuum. Then, the deposition was carried out until required thickness. At last, the samples were post annealed at 140 °C for 2h in air to further improve the crystallinity and opto-electronic properties.

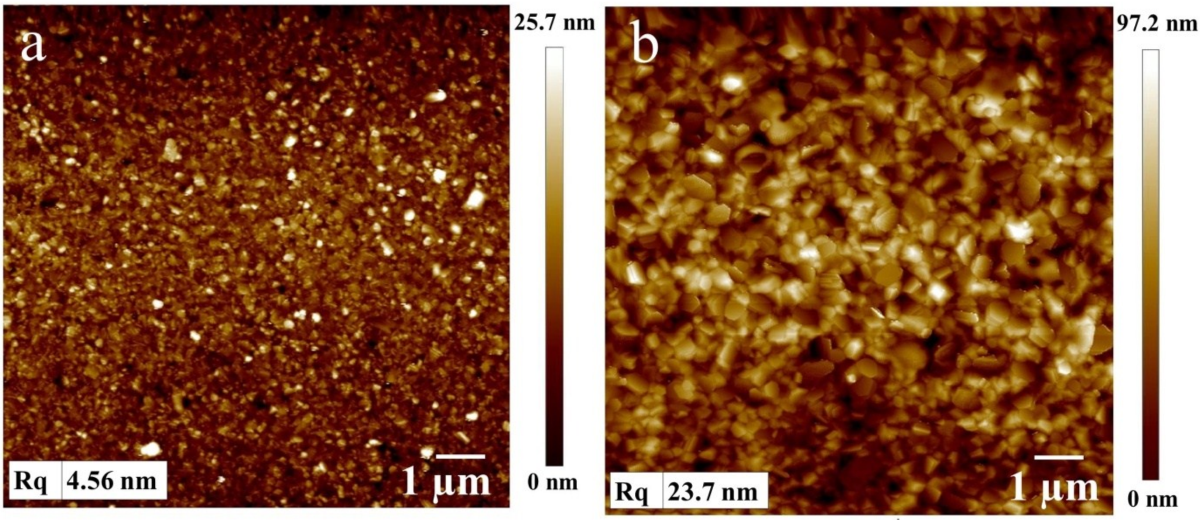


***Fig. S1*** *Comparison of the grain sizes of (a) thermal evaporated and (b) spin-coated perovskite films via high-resolution AFM images.*

## S2. Fabrication of Perovskite Microdisks Using Femtoscond Laser

The parameters we set are the percent of laser power (P) and number of laser direct writing (N) ($P_{max}$≈473 mW). **Fig. S2a** shows a top-view of the microdisks with a diameter of 20 µm patterned at different P and N on a 570 nm thick $FAPbI_3$ film. From **Fig. S2b**, we can see that with the increase of P, the diameter of the microdisks decreases. As **Fig. S2c** shown, we can also find that N does not influence the diameter too much. However, a certain number of cycles are needed to fully pattern thick perovskite films when the percent of laser power is lower than 1%, which could lead to unclear boundaries. By all those concerns, we choose P=1% and N=3 (label 3-3) as the optimal parameter for laser printing of perovskite microdisks. The processing details and the setup can be found in Supporting Information S2.

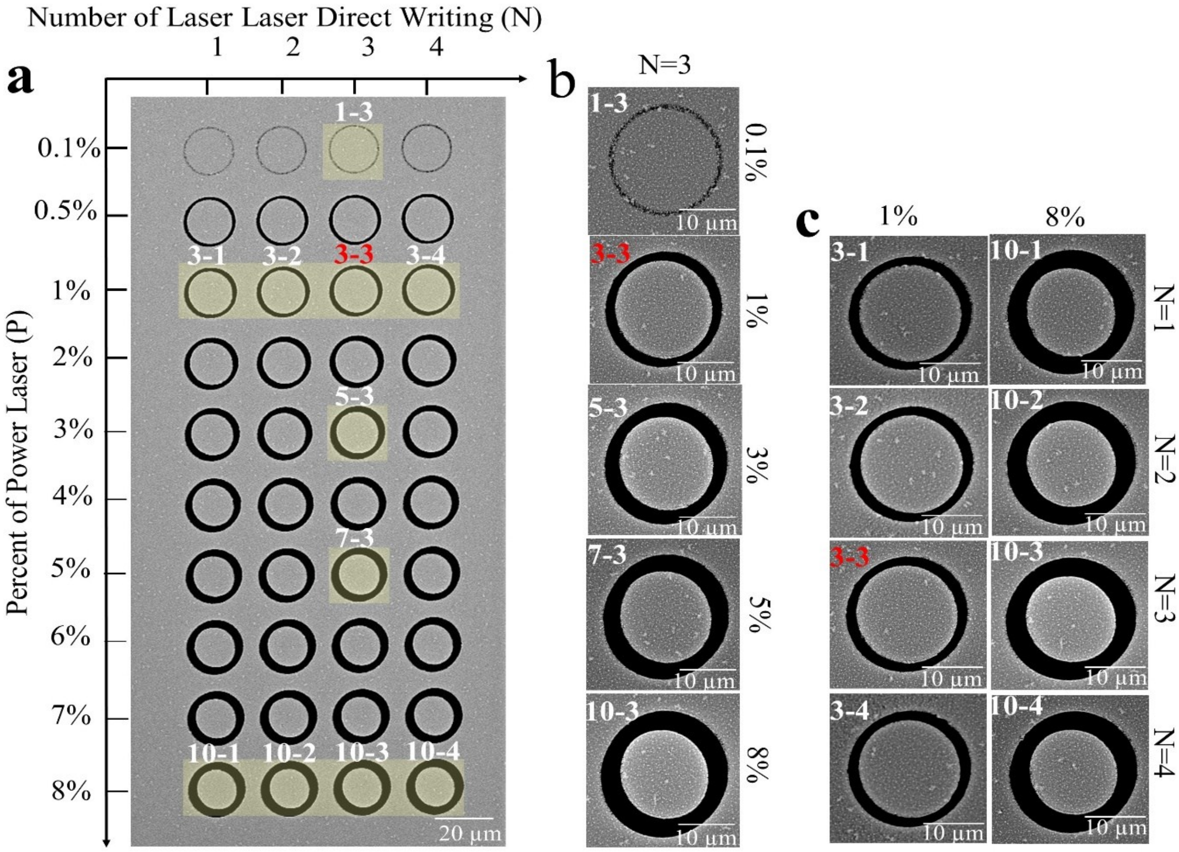


*Fig. S2 (a) SEM image of the perovskite microdisks printed by fs laser with gradually increasing percent of laser power (vertical axis) as well as the processing number of laser writing (horizontal axis), SEM images illustrate several representative surface morphology produced (b) at various P and N= 3, and (c) at various N and P= 1%, P=8%.*

## S3. Scheme of the optical setup for Lasing measurements

Lasing performance of single microdisk was studied under the optical excitation at a wavelength of 532 nm using the combination of Yb: KGW femtosecond laser (Carbide, Light Conversion) and optical parametric amplifier (Orphrus-hp, Light Conversion), yielding laser pulses with a repetition rate of 5 kHz and pulse duration shorter than 267 fs. The laser beam was focused onto the sample surface at back incidence by a 50× microscope objective aligned to ensure uniform irradiation of the disks (Gaussian distribution with an FWHM of 7.1 μm). Emission of a single disk was collected in transmission configuration using the same microscope objective and analyzed with Raman spectrometer (Lab RAM HR Evolution, HORIBA) equipped with a deep cooled open electrode 1024 × 256 CCD camera (Syncerity™ 1024 x 256). The emission of a single disk above the lasing threshold was spatially and spectrally resolved spectrometer using 1800 grooves/mm grating and a pinhole width of 30 μm at the entrance, allowing for 0.65 $cm^{-1}$ spectral resolution. All measurements were performed at room temperature in air. **Fig. S3** is the scheme of the optical setup for micro photoluminescence and lasing measurements.

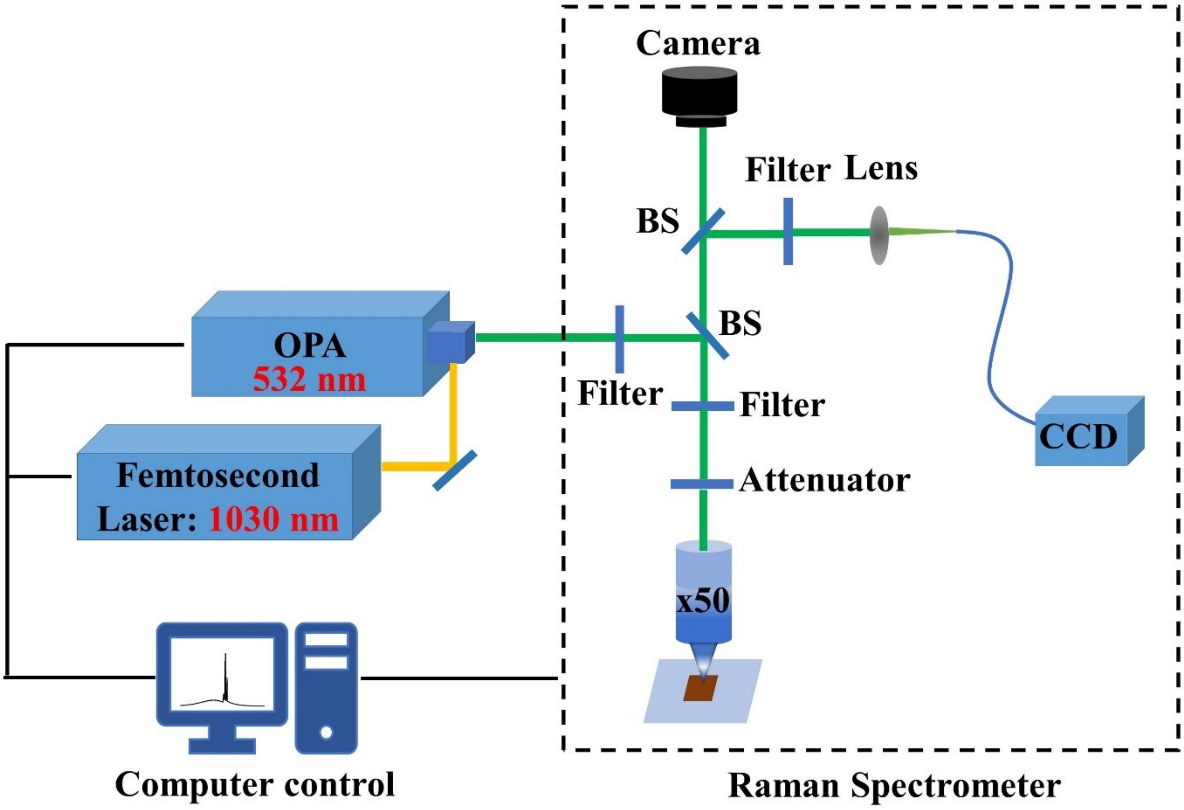


***Fig. S3.*** *Scheme of the optical setup for micro photoluminescence and lasing measurements****.***

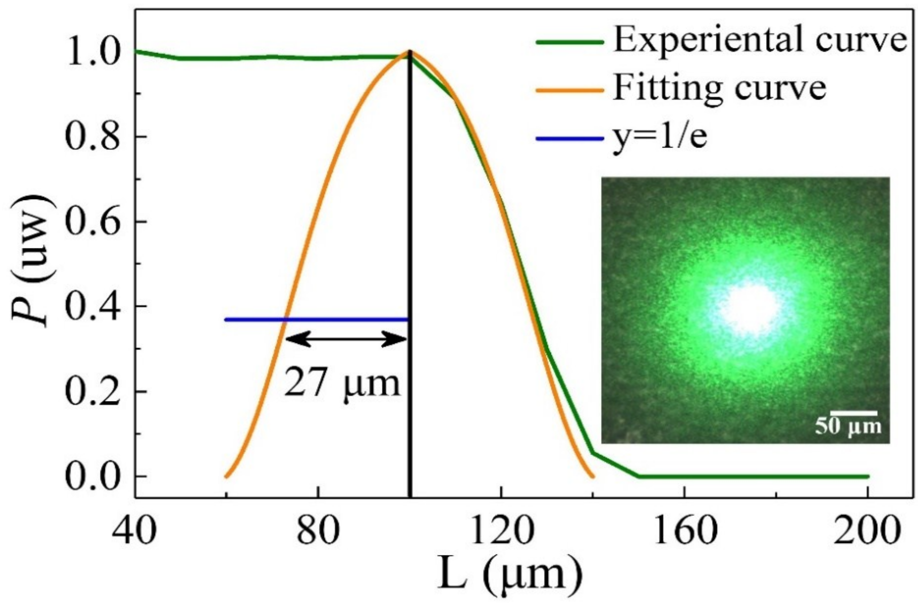


***Fig. S4*** *Calibration of the spot size of laser beam.*

## S4. Lasing Performance of The Perovskite Microdisks with Different Thicknesses

This section provided the microdisks laser performance. Emission spectra of perovskite microdisks with different diameters from 16 μm to 36 μm as shown in **Fig. S5**. The evolution of emission spectra at various pump intensity in different thickness, as seen in **Fig. S6-Fig. S10.**

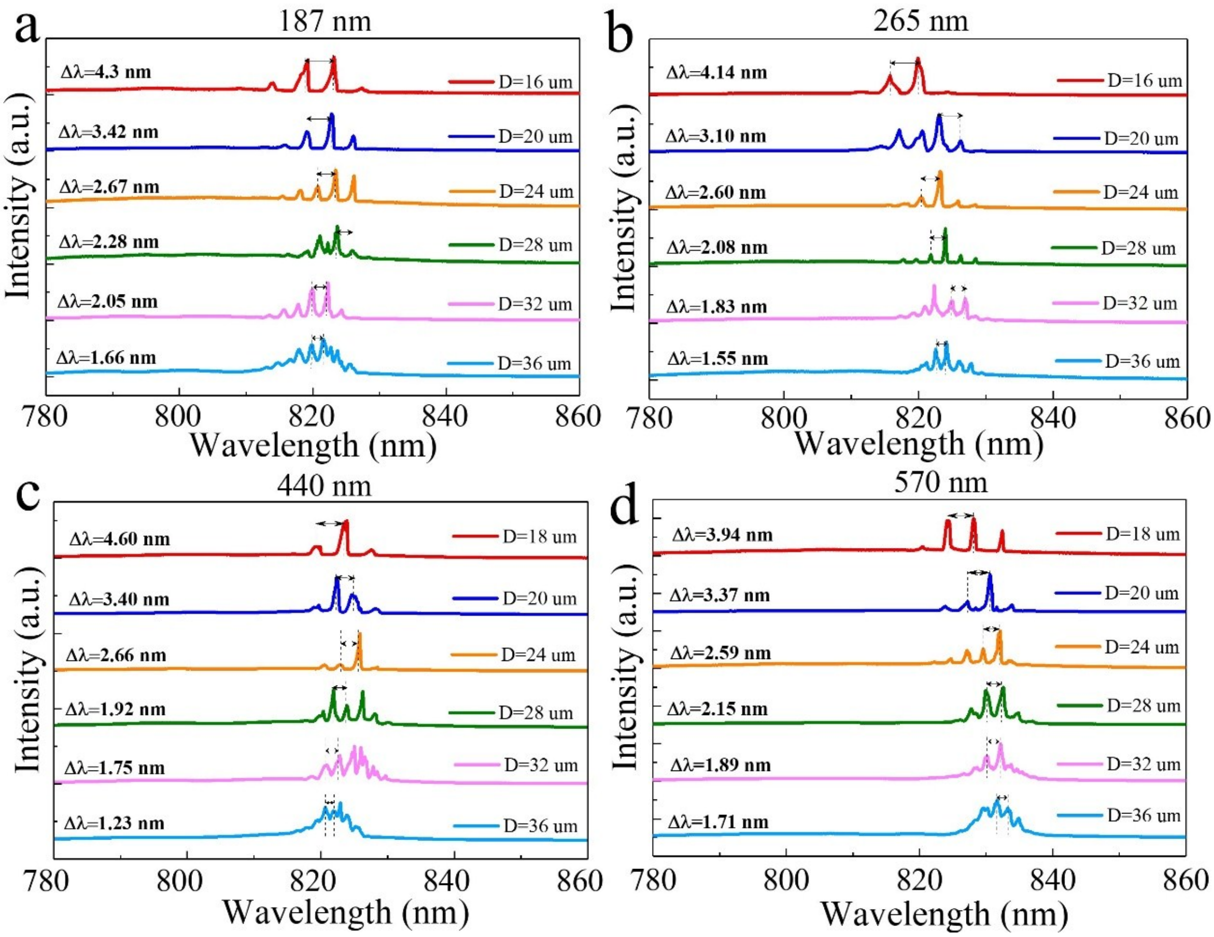


***Fig. S5.*** *Emission spectra of perovskite microdisks with different thicknesses (a) 187 nm, (b) 265 nm, (c) 440 nm and (d) 570 nm.*

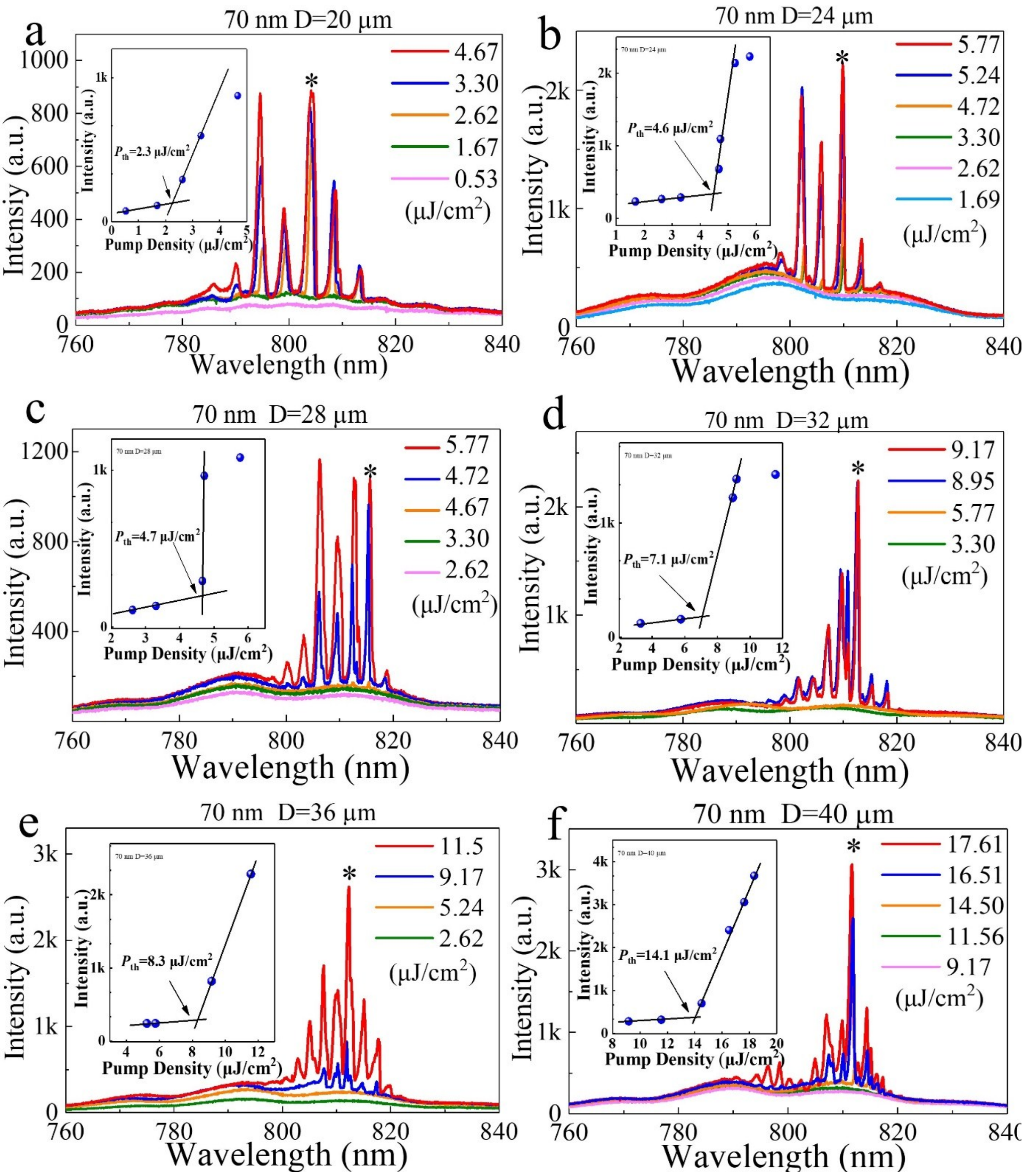


***Fig. S6*** *Emission spectra of poerovskite microdisk with a thickness of 70 nm at different intensities. Inset: PL intensity as a function of pump density. (a) D=20 μm, b) D=24 μm, (c) D=28 μm, (d) D=32 μm, (e) D=36 μm and (f) D=40 μm.*

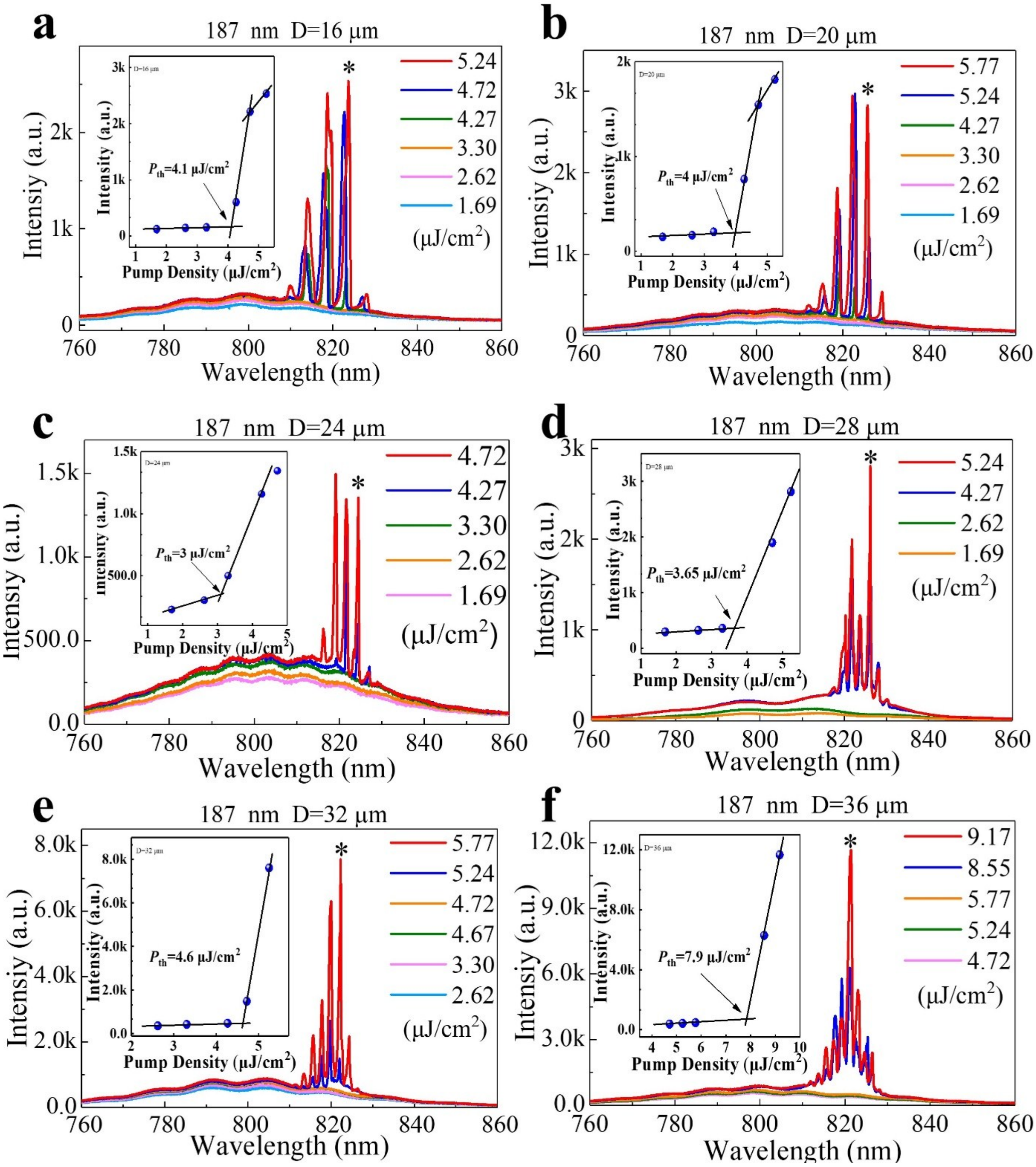


***Fig. S7** Emission spectra of poerovskite microdisk with a thickness of 187 nm at different intensities. Inset: PL intensity as a function of pump density. (a) D=16 μm, (b) D=20 μm, (c) D=24 μm, (d) D=28 μm, (e) D=32 μm and (f) D=36 μm.*

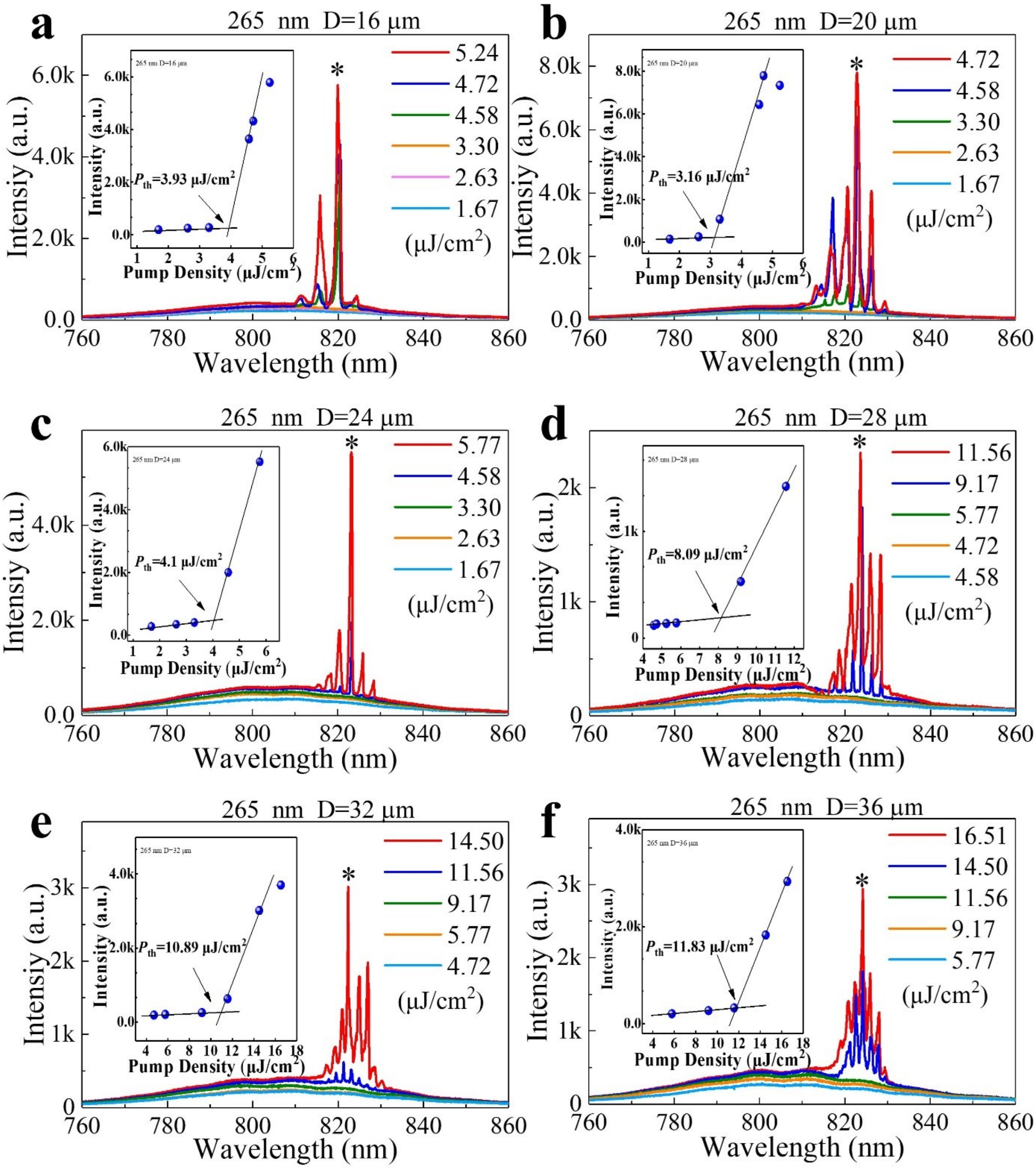


***Fig. S8** Emission spectra of poerovskite microdisk with a thickness of 265 nm at different intensities. Inset: PL intensity as a function of pump density. a) D=16 μm, b) D=20 μm, c) D=24 μm, d) D=28 μm, e) D=32 μm and (f) D=36 μm.*

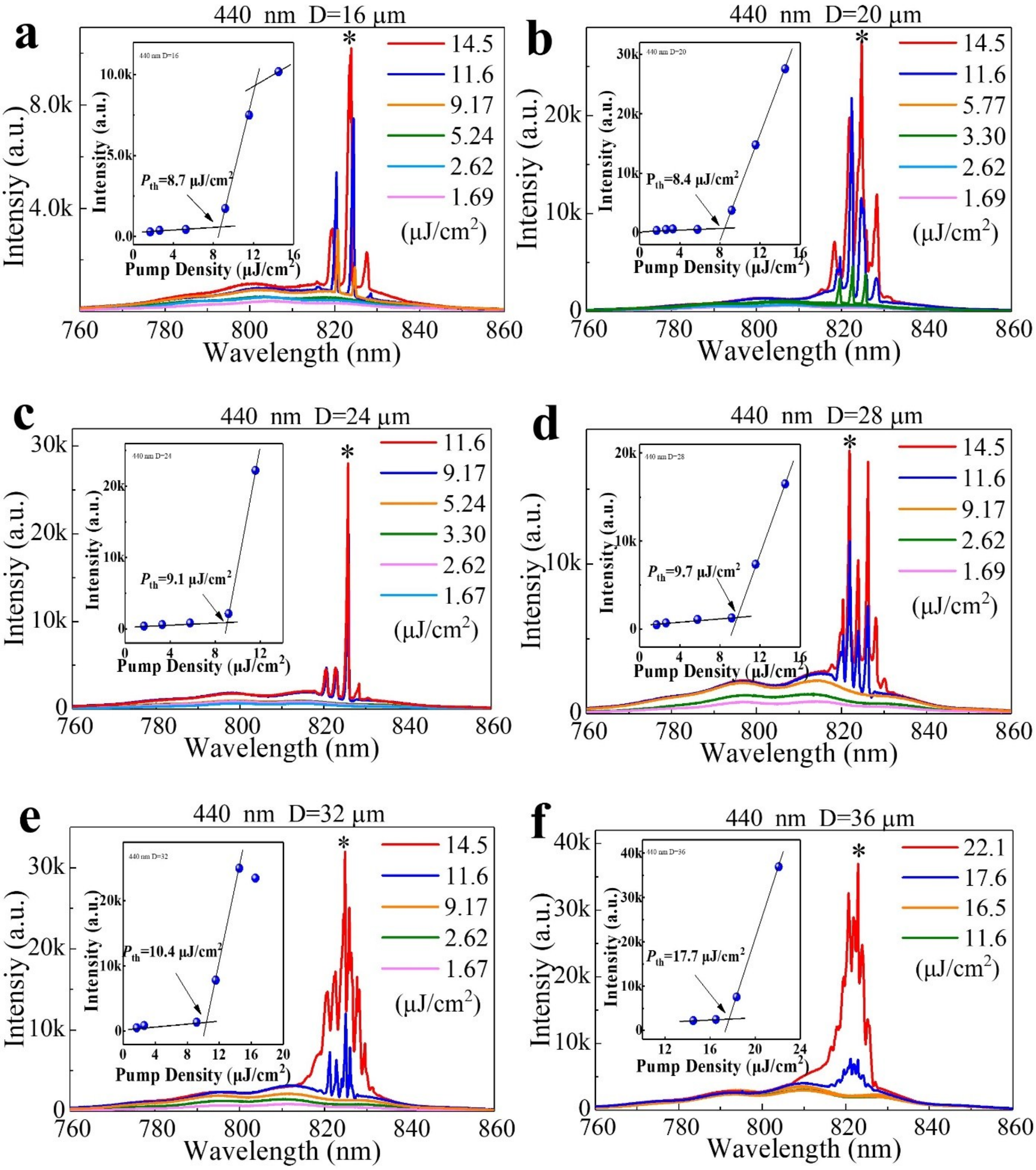

***Fig. S9*** *Emission spectra of poerovskite microdisk with a thickness of 440nm at different intensities. Inset: PL intensity as a function of pump density. (a) D=16 μm, (b) D=20 μm,(c) D=24 μm, (d) D=28 μm, (e) D=32 μm and (f) D=36 μm*

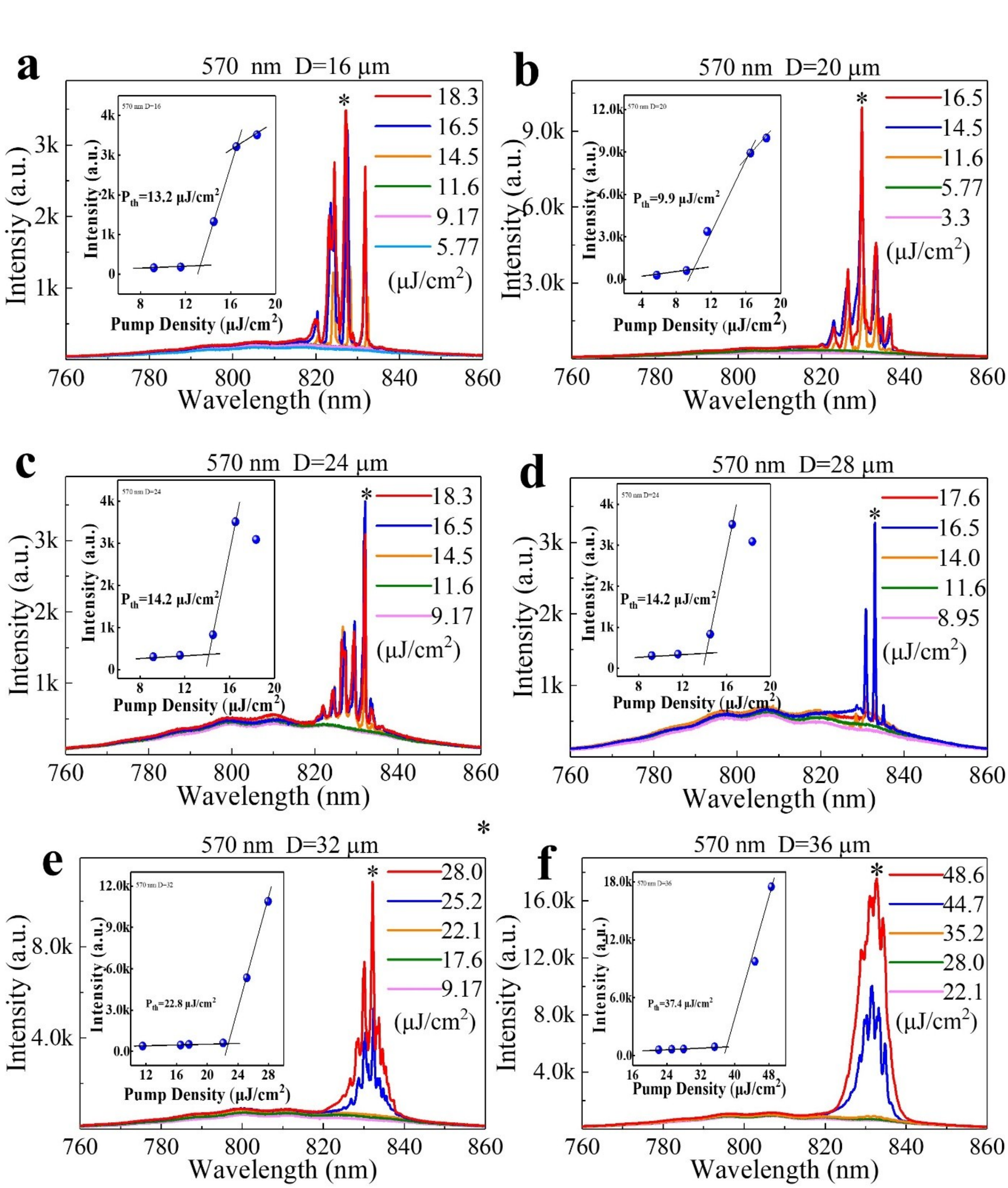


***Fig. S10*** *Emission spectra of poerovskite microdisk with a thickness of 570 nm at different intensities. Inset: PL intensity as a function of pump density. (a) D=16 μm, (b) D=20 μm, (c) D=24 μm, (d) D=28 μm, (e) D=32 μm, (f) D=36 μm*

## S5. Transient Absorpyion Spectra of Perovskite Films with Different Thicknesses

In this section, we test the transient absorption (TA) spectra of $FAPbI_3$ films with different thicknesses, as shown in **Fig. S11**. Combined Yb: KGW femtosecond laser (Carbide, Light Conversion) with optical parametric amplifier (Orphrus-hp, Light Conversion), we got the 532 nm fs-laser pulses which repetition rate is 50 kHz and pulse duration shorter than 267 fs. Ultrafast transient absorption spectra were recorded with a commercial spectrometer (HARPIA-TA, Light Conversion).

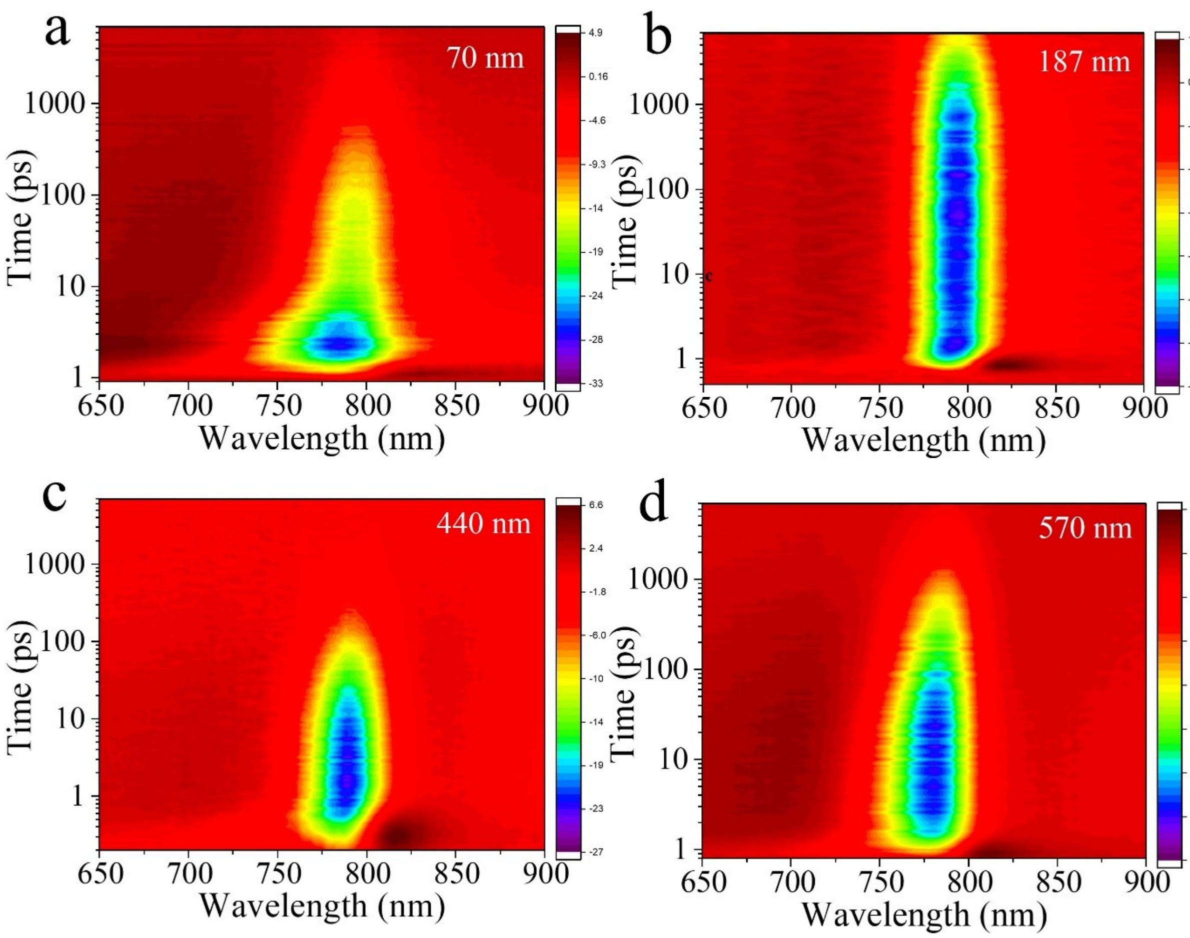


***Fig. S11*** *Transient absorption spectra of $FAPbI_3$ films with different thicknesses. (a) 70 nm, (b) 187 nm, (c) 440 nm, and (d) 570 nm.*