SUPPORTING INFORMATION

Near-infrared random lasing realized in solution-processed perovskite thin film

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Investigation on the phase coexistence phenomenon of CH₃NH₃PbI₃ thin film

As shown in Figure S1, we performed the temperature-dependent XRD measurements at three typical temperature points (300, 160, and 77 K) to support our findings from PL spectra. At 300 K, the diffraction peaks at 14.35°, 28.66°, and 43.18° could be assigned to the (110), (220), and (330) planes of crystalline CH₃NH₃PbI₃, suggesting the formation of a tetragonal perovskite structure (space group *I*4/*mcm*). At 160 K (structural phase transition temperature stated in the manuscript), some emerging diffraction peaks at 14.19°, 28.36°, 31.62°, and 41.10° can be observed, corresponding to (101), (202), (301), and (242) diffractions of orthorhombic structure. Obviously, the existence of residual tetragonal phase implies an incomplete phase transition of CH₃NH₃PbI₃ product. More importantly, such a phase coexistence phenomenon still exists although the measurement temperature is as low as 77 K. The temperature-dependent XRD results match well with the findings in temperature-dependent PL shown in the manuscript.



Figure S1. XRD patterns of the CH₃NH₃PbI₃ thin film at three typical temperatures. The asterisk is used to identify the appearance of orthorhombic phase.

Gaussian deconvolution of the PL spectra at different temperatures

Low-temperature photoluminescence (PL) at three typical temperature points were put together for a better comparison because the carrier recombination processes were changed with temperature. As shown in Figure S2, three PL spectra at 77, 90, and 100 K could be resolved into three components, centered at around 748, 780, and 811 nm, respectively. The component on the high-energy side (~748 nm, R-1) is attributed to the low-temperature orthorhombic phase. The medium component at ~780 nm (R-2) is associated with the high-temperature tetragonal phase. And the component on the low-energy side (~811 nm, R-3) can be ascribed to the trap-mediated radiative recombination. Obviously, with the decrease of temperature, the contribution of carrier recombination channel from R-1 increases gradually, while the opposite is the case for R-2. Specifically, the relative percentages (η %) of R-1 for the PL spectra at three temperature points are 12.25%, 7.79%, and 5.27%, respectively, as summarized in Table 1.



Figure S2. Gaussian deconvolution of the PL spectra measured at (a) 77, (b) 90, and (c) 100 K,

respectively.

<u>Relative percentages of three recombination channels in CH₃NH₃PbI₃ film at <u>different temperatures</u></u>

Temperature	Parameters	R-1	R-2	R3
77 K	Wavelength (nm)	749	782	812
	η %	12.25	30.40	57.35
90 K	Wavelength (nm)	748	779	807
	η %	7.79	34.87	55.34
100 K	Wavelength (nm)	746	778	779
	η %	5.27	40.37	54.36

Table S1. Detailed data from the Gaussian deconvolution of PL spectra at different temperatures

Analysis of the carrier recombination mechanisms of CH₃NH₃PbI₃ thin film

The PL spectrum of CH₃NH₃PbI₃ thin film at 77 K was taken as the research object to further investigate its carrier recombination mechanism. As shown in Figure S3, the PL spectrum

can be well-fitted by three Gaussian peaks, and each emission band corresponds to a particular recombination process, as described above.



Figure S3. Gaussian deconvolution of the PL spectrum measured at 77 K showing three different carrier recombination channels.

Lineshape analysis of CH₃NH₃PbI₃ thin film at 77 K

As shown in Figure S4, the steady-state PL spectrum measured at room-temperature with the excitation power of 3.0 mW can be fitted with two components, exciton-related emission

(Gaussian, blue) plus free carrier-related emission (Gaussian, green). In addition, the ratio of free carrier-related emission to exciton-related emission is calculated to be \sim 7.2%.



Figure S4. Gaussian deconvolution of the steady-state PL spectrum measured at roomtemperature with the excitation power of 3.0 mW, showing the exciton-related emission and free carrier-related emission.

Excitation power-dependent PL intensity of CH₃NH₃PbI₃ thin film at 170 K

Figure S5 shows the relationship between the integrated PL intensity and the excitation power measured at 170 K. The obtained data was fitted by the equation of $I_{PL} = I_{EX}^{\beta}$, where β denotes the nonlinear component. By fitting the experimental data, a superlinear relation with $\beta \sim$ 1.20 held the curve, smaller than the value (~1.29) derived at room-temperature.



Figure S5. The relationship between the integrated PL intensity and the excitation power

measured at 170 K.