SUPPORTING INFORMATION

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

Zhi-Feng Shi,*a Xu-Guang Sun,a Di Wu,a Ting-Ting Xu,a Yong-Tao Tian,a Yuan-Tao Zhang,b Xin-Jian Li*a and Guo-Tong Du*b

aDepartment of Physics and Laboratory of Material Physics, Zhengzhou University, Daxue Road 75, Zhengzhou 450052, China

bState Key Laboratory on Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, Qianjin Street 2699, Changchun 130012, China

*Author to whom correspondence should be addressed. Electronic mail: shizf@zzu.edu.cn; lixj@zzu.edu.cn
Investigation on the phase coexistence phenomenon of CH$_3$NH$_3$PbI$_3$ 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 CH$_3$NH$_3$PbI$_3$, suggesting the formation of a tetragonal perovskite structure (space group $I4/mcm$). 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 CH$_3$NH$_3$PbI$_3$ 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.

![XRD patterns of the CH$_3$NH$_3$PbI$_3$ thin film at three typical temperatures. The asterisk is used to identify the appearance of orthorhombic phase.](image)

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.

**Relative percentages of three recombination channels in CH$_3$NH$_3$PbI$_3$ film at different temperatures**
<table>
<thead>
<tr>
<th>Temperature</th>
<th>Parameters</th>
<th>R-1</th>
<th>R-2</th>
<th>R3</th>
</tr>
</thead>
<tbody>
<tr>
<td>77 K</td>
<td>Wavelength (nm)</td>
<td>749</td>
<td>782</td>
<td>812</td>
</tr>
<tr>
<td></td>
<td>$\eta$ %</td>
<td>12.25</td>
<td>30.40</td>
<td>57.35</td>
</tr>
<tr>
<td>90 K</td>
<td>Wavelength (nm)</td>
<td>748</td>
<td>779</td>
<td>807</td>
</tr>
<tr>
<td></td>
<td>$\eta$ %</td>
<td>7.79</td>
<td>34.87</td>
<td>55.34</td>
</tr>
<tr>
<td>100 K</td>
<td>Wavelength (nm)</td>
<td>746</td>
<td>778</td>
<td>779</td>
</tr>
<tr>
<td></td>
<td>$\eta$ %</td>
<td>5.27</td>
<td>40.37</td>
<td>54.36</td>
</tr>
</tbody>
</table>

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

**Analysis of the carrier recombination mechanisms of CH$_3$NH$_3$PbI$_3$ thin film**

The PL spectrum of CH$_3$NH$_3$PbI$_3$ 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 CH$_3$NH$_2$PbI$_3$ 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 ~7.2%.

![Gaussian deconvolution of the steady-state PL spectrum](image)

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

**Excitation power-dependent PL intensity of CH$_3$NH$_3$PbI$_3$ 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 $\beta$
denotes the nonlinear component. By fitting the experimental data, a superlinear relation with $\beta \sim 1.20$ held the curve, smaller than the value ($\sim 1.29$) derived at room-temperature.

Figure S5. The relationship between the integrated PL intensity and the excitation power measured at 170 K.