

[Supplementary File](#)

**Artificial intelligence-assisted pump-locked random lasers**

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## 1. Design and fabrication process of random lasers.

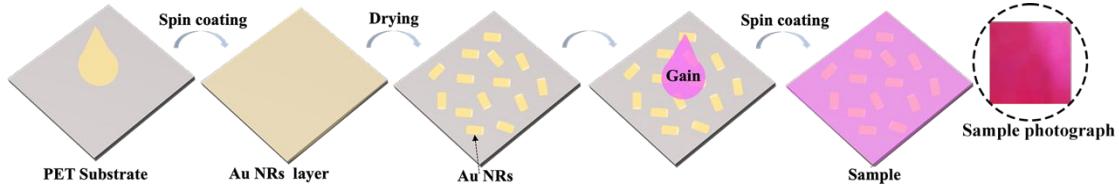


Fig. S1 Sample preparation process and sample photograph.

The disordered system is a polymer film fabricated by dye-doped PMMA with scattering nanoparticles. Herein, a typical laser dye RhB is used as gain material, while Au NRs with diameter and length about 25 and 50 nm act as scatterers. The fabrication process is illustrated in Fig. S1. Firstly, the Au NRs (0.02 mg/mL) was spin-coated on the flexible PET substrate at a speed of 1800 r/min for 40s. Secondly, the RhB (6 mg/mL) and PMMA (200 mg/mL) were mixed with a volume ratio of 1:1 under magnetic stirring for 20 min. Then, spin coating the mixture on the Au NRs layer at a speed of 1800 r/min for 40 s. Thirdly, heat the structure at 70 °C for 20 min for solidification. Finally, the disordered system is obtained, which is uniform as shown in sample photograph. The solvent here was dichloromethane.

## 2. Schematic diagram of mode selection of random lasers using our intelligence control system.

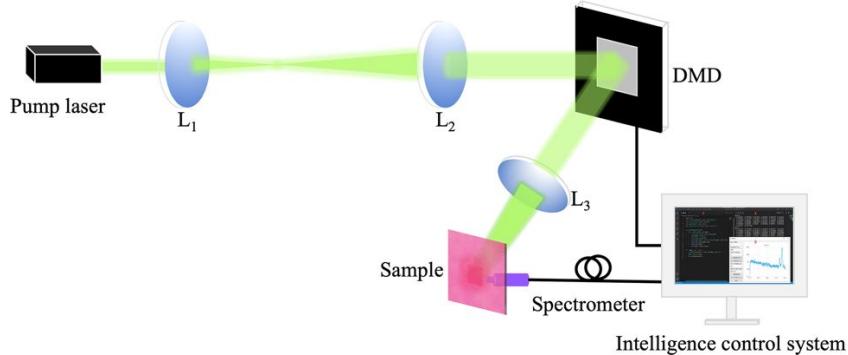


Fig. S2 The diagram of experimental setup for mode selection using intelligent control system.

A doubled Q-switched Nd:YAG laser (532 nm, 5-7 ns, 10 Hz) is employed as the pump source. The lens 1( $L_1$ ) and lens 2 ( $L_2$ ) are used for expanding and collimating of pump beam, respectively. The DMD is used to shape the pump profile based on the feedback of grayscale of image. The modulated pump light is reflected to lens 3 ( $L_3$ ). The lens 3 is used to adjust the size of pump profile. The modulated pump pattern incident on the sample and the reflected emission light was collected by an optical fiber spectrometer (Ocean Optics HR4000) with a spectral resolution of 0.02 nm. The pump power was controlled by pump voltage. The pump source, digital micromirror device (DMD) and high-resolution spectrometer are linked using the self-development software.

### 3. The influence of fiber probe position on laser modes

Based on the optimized pump profile of the single-mode at 589 nm (Figure 2 in the manuscript), we have studied the influence of fiber probe position on laser modes. Once the pump profile is fixed, the emission spectra have been collected with fixed pumping energy density (1.51 MW/cm<sup>2</sup>) at various observation angles ( $\theta = -60^\circ, 30, 0, -30, -60^\circ$ ,  $\theta$  represents the angle formed by the normal vector of the sample and the direction of observation), as shown in Figure R1. The result indicates the lasing characteristics (wavelength, the line width, and the intensity) are almost no change, which is a distinct indication of the occurrence of random laser activity. That is to say, the laser modes are independent of detection position.

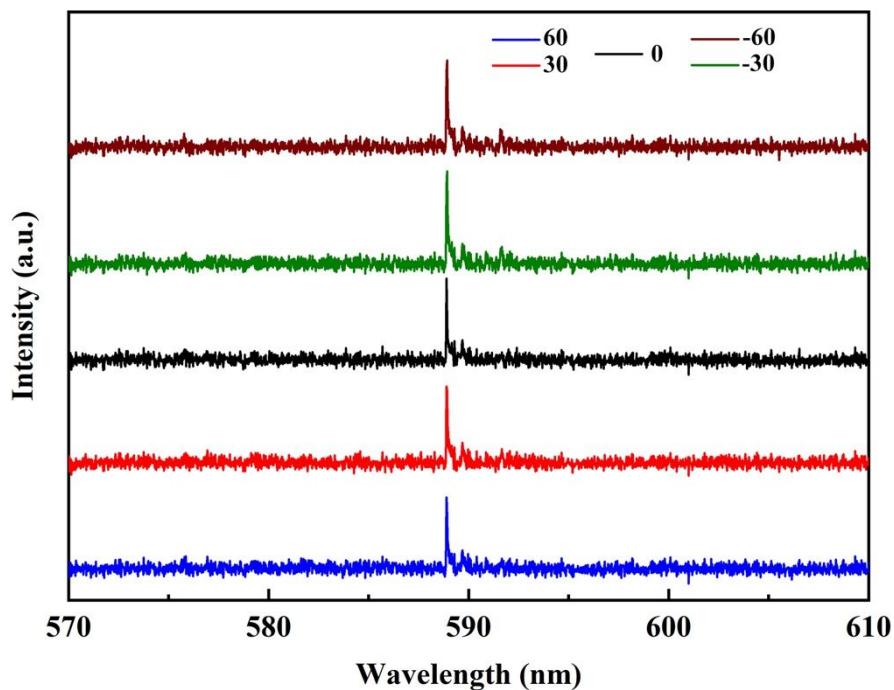


Fig. S3 Emission spectra of optimized random laser at various detection angles with the fixed pumping energy density (1.51 MW/cm<sup>2</sup>)

### 4. Extraction of arbitrary single-mode.

In our experiment, the modes ② and ③ can also be individually extracted effectively. We have added the optimized results of modes ② and ③, as shown in Figure R2, which present the lasing spectra and corresponding optimized pump profiles of optimized modes ③ and ②.

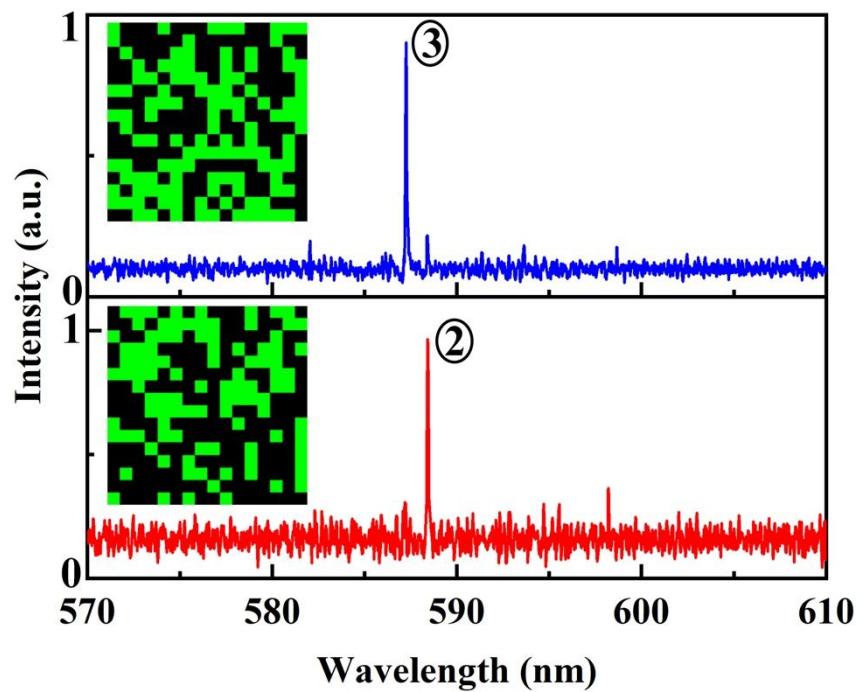


Fig.S4 Random lasing spectra and the corresponding optimized pump profiles with a single mode

at 587.0 nm and 588.1 nm (modes ③ and ②)

**5. The optimization process for target modes in Figure 4. The corresponding movies (movies S1 to S3) have also uploaded.**

**Movie S1:** The optimization process for single mode at 591.5 nm ( $\lambda_1$ )

**Movie S2:** The optimization process for single mode at 594.0 nm ( $\lambda_2$ )

**Movie S3:** The optimization process for two modes ( $\lambda_1$  &  $\lambda_2$ ) simultaneously