Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

A ring-shaped random laser in momentum space

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A: SEM image of TiO_2 nanoparticles in polymer film

TiO_2 nanoparticles are used as the scatterer to provide effective feedback for random system as shown in Fig. S1. It can been seen that some TiO_2 nanoparticles are randomly distributed in the polymer film. And some TiO_2 nanoparticles are aggregated into larger particles in the scale of few hundreds nanometers as shown the insert in Fig.S1. These TiO_2 nanoparticles including the aggregating state supply random optical feedback for the gain materials to achieve the stimulated emissions.

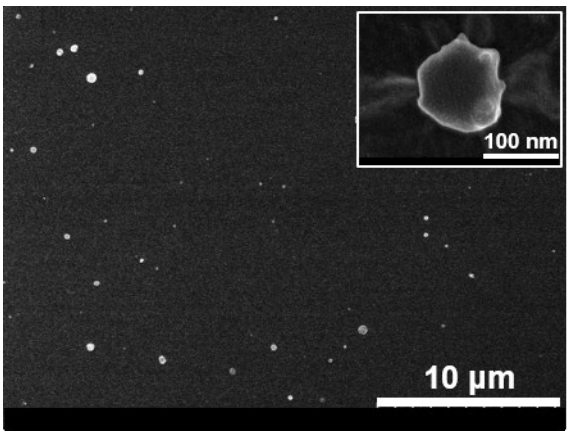

Fig. S1. SEM image of TiO_2 nanoparticles in the polymer film.

B: Emission angle range of random lasing from fiber ports

When the dye-doped polymer film is pumped, random lasing can be emitted from the random film with the refraction index $n_1 = 1.4$ when the total gain is larger than the loss. And part of the emission will be coupled into the optical fiber and then propagating along the optical fiber as shown in Fig. S2. For long-distance transmission, the total reflection is required at the interface of the plastic layer with $n_2 = 1.5$ and air. As a result, the refraction angle of θ_2 in the plastic coating should be in the range of $[\arcsin\frac{1}{n_2}, \arcsin\frac{n_1}{n_2}]$ (i.e. $[41.81°, 68.96°]$) for the beam into the fiber with an incidental angle $\theta_1 \in [0°, 90°)$. By using the refraction law, the emission angle γ of the random lasing from the end of plastic layer should be in the range $[32.58°, 90°)$. Similarly, under the condition that the random laser can be totally reflected at the interface between the plastic coating and the air, the output angles γ are greater than 36.30° and 39.91°, when the random lasing is emitted from the end of cladding layer with $n_3 = 1.52$ and the fiber core with $n_4 = 1.54$.

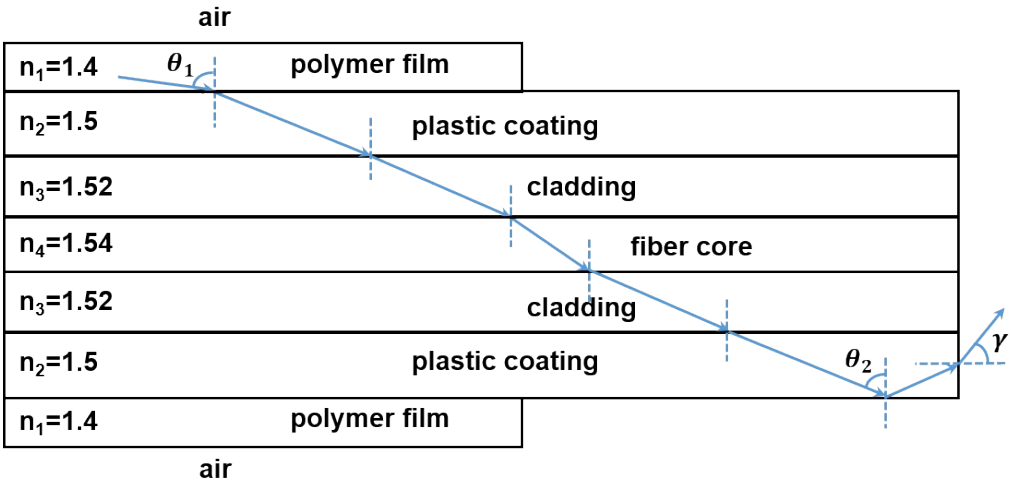

Fig. S2. Schematic diagram of the optical path when a random lasing beam coupled into an optical fiber propagates through the fiber.

C: Emission performances of red random laser coupled with fiber

The influence of the pumping angle φ on the output performance of the random lasers is demonstrated as shown in Fig. S3a. Figure S3b shows the emission spectrum of the random laser measured at different incident angles when the pump power density is 0.6565 MW cm^{-2}. The multi-mode random lasing can be achieved at an extreme wide angle range of [20°, 160°]. Meanwhile, as the pumping angle φ increasing from 20° to 160°, the corresponding threshold remains almost unchanged at a small value of 0.1194 MW cm^{-2} (Fig. S3c). The stability in threshold indicates that the random lasers can be easily obtained for further usage.

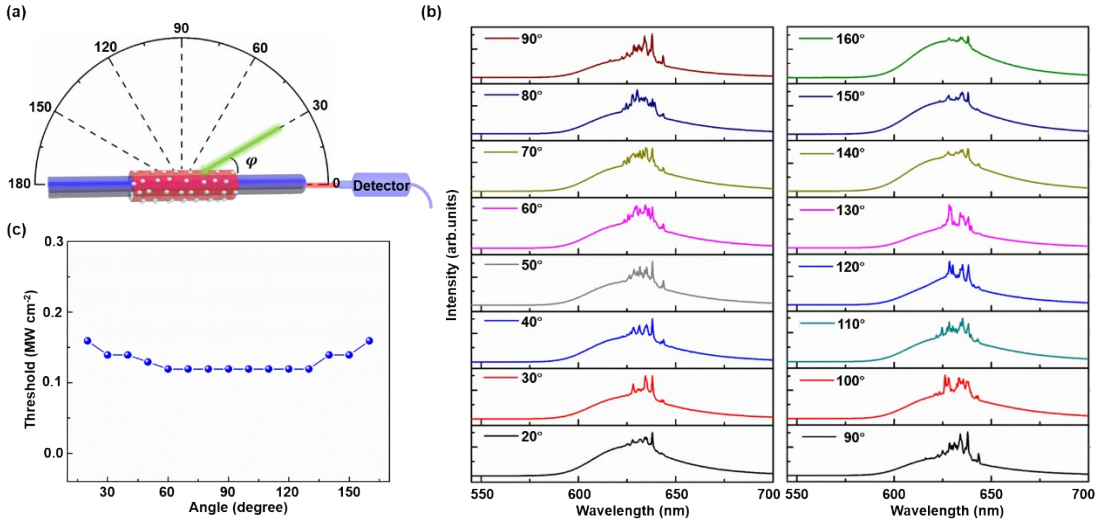

Fig. S3. Effect of the pumping angle φ on the output performance of random lasers. (**a**) Schematic diagram of the detection with different pumping angles. (**b**) The emission spectra of the random laser pumped at different pumping angles when the pump power density is 0.6565 MW cm^{-2}. (**c**) The threshold of the random laser changes with the pumping angle.

The effect of the thickness on the emission spectra of random lasing is also demonstrated in Fig. S4a by maintaining the pump power density at 0.2188 MW cm^{-2}. The intensity of random lasing increases with increasing the film thickness, following a red-shift of the center wavelength about 10 nm. This red-shift phenomenon may be induced by reabsorption of the dye molecules in a thicker gain film. As shown in Fig. 3a, the overlap between the

absorption and emission spectra of the DCJTB molecule means that a portion of the radiant lights are reabsorbed by the DCJTB molecules which are excited and give a emission with a relatively long wavelength in a thicker film. And a thicker film supplies a longer optical path for the emitted photons. As a result, the emission random lasing demonstrates a spectral red-shift phenomenon with increasing the thickness of the film. This result indicates that the emission wavelength of the random laser can be adjusted by changing the thickness of the coating polymer film.

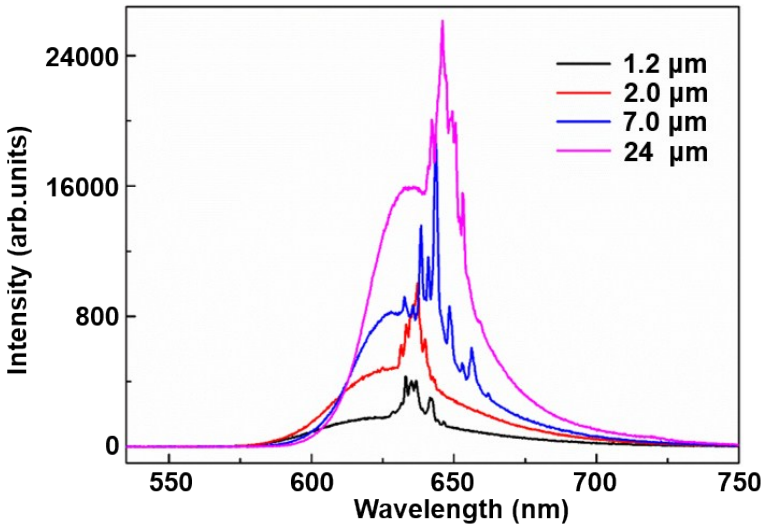

Fig. S4. Emission spectra of the random laser with different thicknesses of polymer film at a pump power density of 0.2188 MW cm^{-2}.

Figure S5a shows the emission spectral variation with the propagating distance *L* in the optical fiber at a pump power density of 7.7190 MW cm^{-2}. Coherent random laser can still be measured even when the propagating distance *L* reaches 20 cm although the intensity of the random lasing decreasing with *L* increasing (Fig. S5b). These experimental results indicate that the fabricated fiber source can be used in an integrated optical system.

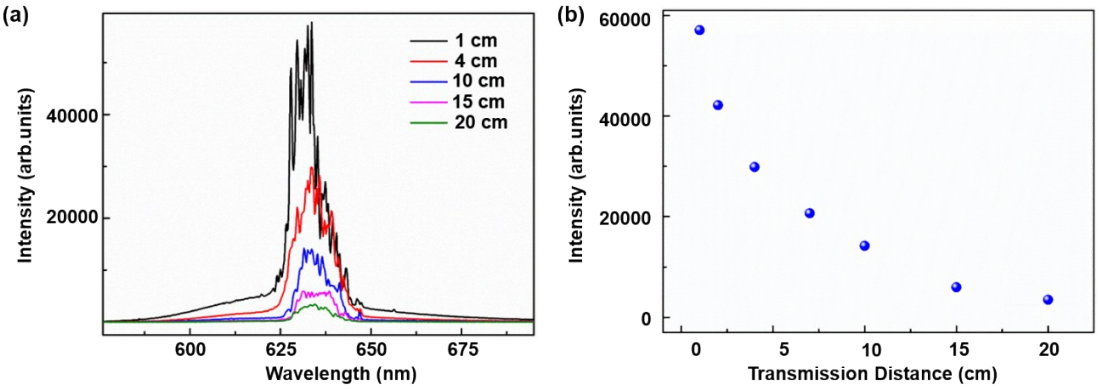

Fig. S5. (**a**) Emission spectra of random lasing through different propagating distances at a pump power density of 7.7190 MW cm^{-2}; (**b**) The peak intensity of random lasing changes with the propagating distance.

Figure S6 shows the emission spectra of the red random laser at different detection angles of α = -30°, -15°, 0°, 15° and 30° at a pump power density of 0.9947 MW cm^{-2}, respectively. Coherent random lasing can be observed at all the detection angles. But the peak intensity of random lasing at α =0° is about 30 times than that of lasing at α = -30°. The large contrast indicating the random lasing is mainly concentrated in the optical fiber, which proves that the fiber source has a good output directionality.

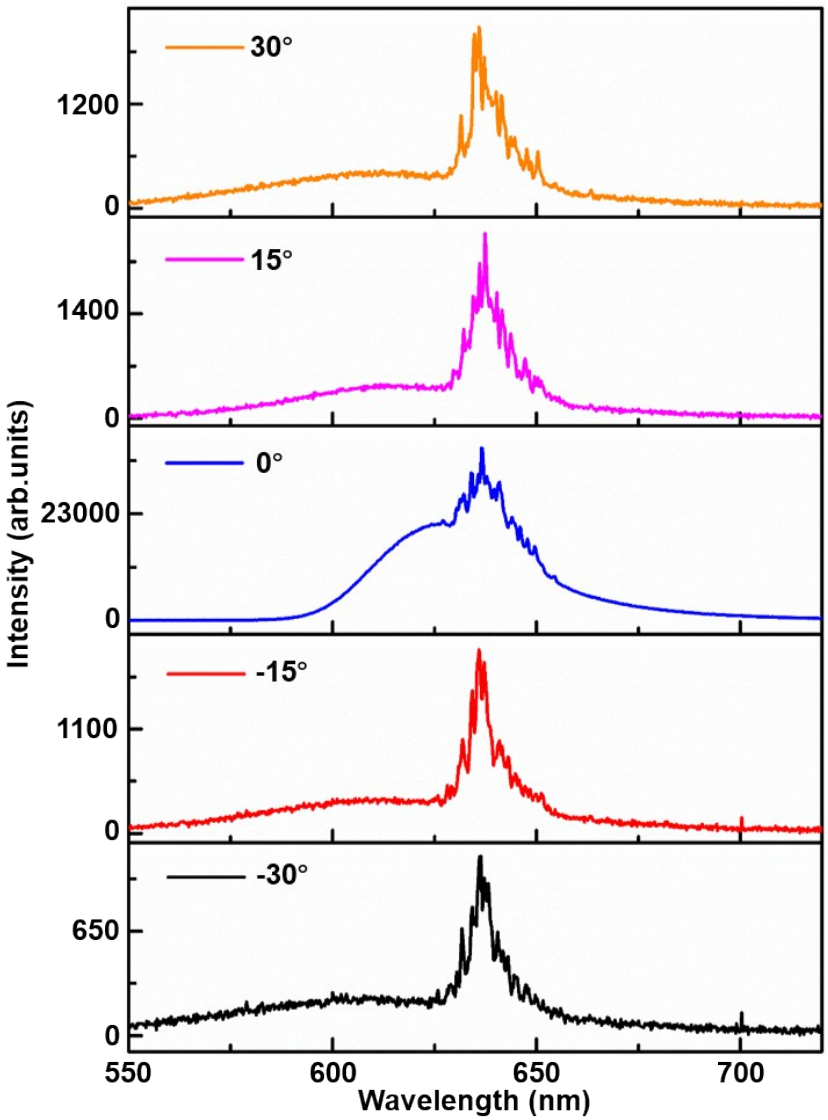

Fig. S6. Emission spectra measured from different detection angles α at a pump power density of 0.9947 MW cm^{-2}.

D: Designing color-switchable random laser

The color of random lasing from the fiber source can be flexibly switched by integrating different random lasers on one optical fiber. A color-switchable random laser is fabricated as an example in Fig. S7a. The two dyes of PM567, DCJTB are separately used as the gain material in the two random lasers, respectively. Figure S7b shows the absorption and photoluminescence spectra of the PM567 film with a dye concentration of 1.25 mg mL^{-1}, demonstrating strong absorption at the exciting wavelength of 532 nm. A triple-state switchable random laser between yellow, red and yellow-red random lasing is thus achieved by simply changing the pump position. When the red random laser with DCJTB is excited alone, red random lasing is achieved as shown in Fig. 3b. And yellow random lasing is obtained as shown in Fig.5b when the yellow random laser with PM567 is pumped alone. Moreover, the dual-color random lasing can be obtained when the two adjacent dye polymer films are simultaneously pumped as shown the inset in Fig. S8. The corresponding emission spectra of the dual-color random laser are shown in Fig. S8 when the sample is pumped at different pump power densities. The red and yellow

coherent random lasing are simultaneously observed as the pump power density is greater than 0.1592 MW cm^{-2}. Therefore, it is concluded that a color-switchable random laser between yellow, red and a yellow-red dual-color output can be easily achieved by mechanically controlling the pump position.

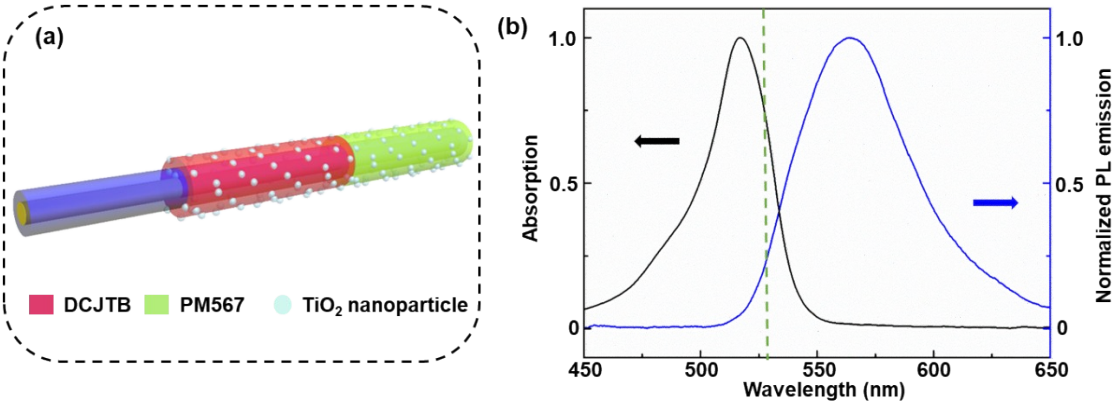

Fig. S7. (**a**) Schematic diagram of the red-yellow switchable fiber source. (**b**) The absorption (black) and photoluminescence (blue) spectra of the PM567 film with a dye concentration of 1.25 mg mL^{-1}.

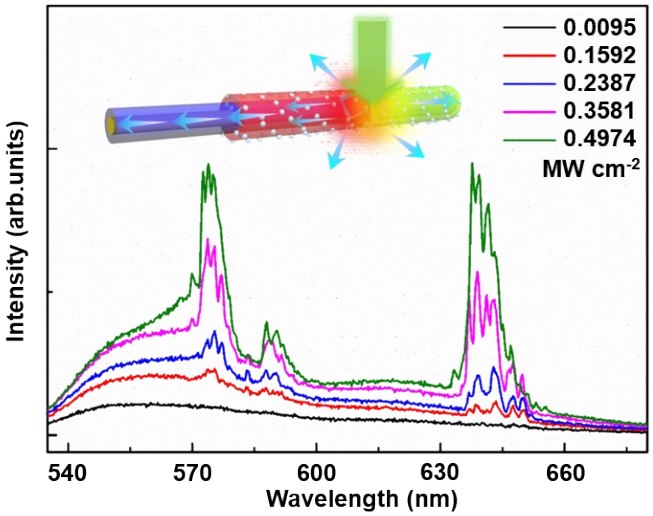

Fig. S8. Emission spectra of the two-color random lasing.