A ring-shaped random laser in momentum space†

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A ring-shaped random laser in momentum space is designed by directly coupling a random laser with a commercial optical fiber. By using a simple approach of selectively coating the random gain layer on the surface of the fiber, red and yellow random lasers are respectively achieved with low threshold values and a good emission direction due to the guiding role of optical fibers. The unique coupling mechanism leads to a random laser with a ring shape in momentum space, which is an excellent illuminating source for high-quality imaging with an extremely low speckle noise. More importantly, a triple-state color-switchable random laser with yellow, red and yellow-red dual-colors can be flexible, and is obtained by simply moving the pump position. The results may promote the practical applications of random lasers in the fields of sensing, in vivo biological imaging, and high brightness full-field illumination.

1. Introduction

In recent years, lasers have been considered as potential illumination sources for the next generation of projectors.1 However, the high spatial coherence of lasers results in coherent imaging artifacts caused by interference that occurs during image formation. To improve their applications in laser display and imaging, some techniques have been utilized to passively decrease the spatial coherence of lasing for suppressing coherent artifacts.2–5 Therefore, the active methods of eliminating the coherent noise are highly desired for the next generation of real-time display technology.

As a new type of laser, random lasers utilize multiple scattering to achieve stimulated radiation amplification, avoiding the complex manufacturing process and high cost of conventional resonators.6–12 Random lasers have become a hot research field in the international laser science community due to their unique characteristics of low cost, small size and ease of integration, demonstrating versatile application prospects in bio-detection,13,14 information security,15 and integrated photoelectron.16 More importantly, the low spatial coherence of random lasing naturally leads to the shortcomings of traditional lasers in full-field imaging and display. Random lasers are considered as ideal illumination light sources for speckle-free full-field imaging.17–22 However, the randomness of the emission direction seriously hinders their practical applications.23,24 Several technical solutions have been proposed to control the emission direction of a random laser by coupling the random laser with an optical fiber or a waveguide structure.25–27 Specifically, the plasmonic random lasers were designed on the fiber end facet28 and in polymer fiber29 to control the emission direction of the random lasing. These fiber sources are receiving more and more attention due to their directionality, long-distance transmission and easy integration. Notably, the output from these light sources is uniform circular spots in the momentum space. Considering the ring-shaped light sources demonstrating great advantages in suppressing coherent noise30 and super-resolution imaging,31 it is necessary to design a ring-shaped random laser to further improve the quality of imaging for a wider range of application requirements. In addition, the implementation of color-regulated random lasers is critical to a real illumination source, and researchers have continually explored ways to regulate the frequency range of random lasers from ultraviolet,32 visible,33 to near-infrared.34 A color-switchable random laser with high integration and easy implementation has still been highly desired in the field of illumination.

In this work, a ring-shaped random laser in momentum space is proposed by selectively coupling a random gain medium with a commercial optical fiber. By coating different gain dye films, the red and yellow random lasers with low thresholds of 0.1335 and 0.1193 MW cm⁻² are achieved, respectively. A red-yellow dual-color random laser can be obtained when the adjacent random lasers are simultaneously pumped. The obtained random laser has the unique characteristics of a small propagation constant along the optical fiber and thus it is ring-shaped in momentum space. Moreover, the color of random lasing can be flexibly switched by simply...
changing the pumping position. As an illumination source, the random laser with a ring-shape in momentum space assures high-quality imaging with an extremely low speckle noise. The results may hasten the practical applications of the random laser in speckle-free full-field imaging, laser displays, bio-sensing and integrated optics.

2. Experimental methods

The fabrication process of the fiber source is illustrated in Fig. 1a. The typical gain materials used in this experiment are as follows: 4-(dicyanomethylene)-2-tert-butyl-6-(1,1,7,7-tetramethyljulolidin-4-yl-vinyl)-4H-pyran (DCJTB, Tokyo Chemical Industry) and Pyrromethene567 (PM567, Sigma). The fabrication process of the fiber source is performed as follows: first, polydimethylsiloxane (PDMS, n = 1.41) solution is mixed with cross-linking solution in a ratio of 1:10. Then, TiO2 nanoparticles are dispersed in the dye-doped acetone solution (DCJTB at 7.5 mg mL⁻¹ or PM567 at 6.25 mg mL⁻¹) to obtain TiO2 dispersion with a concentration of 1.5 mg mL⁻¹. The dye-doped TiO2 dispersion and PDMS are mixed at a volume ratio of 1:5 in an ultrasonic tank for 15 min and then placed in a vacuum for 40 min to remove the air bubbles. Finally, the mixture is dipped into a clean fiber (TAIHAN Fiber Optics) to flow naturally. The fiber length is about 60 mm, the core diameter is 50 μm and the diameter of the cladding layer is 125 μm. The refractive indexes of the core and cladding layer are 1.54 and 1.52, respectively. The sample is placed in a drying oven at 80 °C for 3 hours to complete the cross-linking polymerization and drying. After cooling to room temperature, a fiber source is realized. Random lasers with different colors can be fabricated by coating different dye polymers at different locations on the surface of the fiber. The polymer film with different thicknesses can be fabricated by controlling the dipping times.

3. Results and discussion

The optical photograph, micrograph and scanning electron microscopy (SEM, Hitachi SU8010) images of the obtained samples are shown in Fig. 1b–e, respectively. The polymer film with a thickness of about 1.2 μm is relatively uniformly coated on the fiber surface. In this film, the TiO2 nanoparticles with a diameter of around 50 nm as shown in Fig. 1f are used as scatterers. Actually, some TiO2 nanoparticles are aggregated into larger particles in the scale of few hundreds of nanometers as shown in Fig. S1 (ESI†). These TiO2 nanoparticles can provide effective feedback for random systems since their refractive index is much larger than the refractive index of PDMS.35,36

The experimental setup for the random laser is shown in Fig. 2a. The sample (S) is mounted on a stage and optically pumped by a frequency-doubled and Q-switched neodymium doped yttrium aluminum garnet (Nd:YAG) laser with a wavelength of 532 nm, 10 Hz repetition rate, and 8 ns pulse duration passing through a half wave plate (HWP) and a Glan prism (GP) for easily changing the pump power density. Then the pulses are split into two beams by using a neutral beam splitter (NBS). One beam is measured by using an optical power meter (OPM) for monitoring the pump power density, and the other beam is used to pump the sample through three mirrors (M1, M2 and M3) for controlling the pumping position of the fiber sample (the inset of Fig. 2a) to achieve the emission of random lasing. The spot size of the pulsed laser is fixed at 8 mm in diameter. The emission spectra are recorded by using a spectrometer of Ocean Optics model Maya Pro 2000 with a spectral resolution of 0.4 nm. All the experiments are performed at room temperature. In our experiments, the detector is approximately 25 mm from the pump position for recording the lasing spectra unless otherwise specified.

The designing mechanism of the ring-shaped random laser in momentum space is schematically demonstrated in Fig. 2b. When the DCJTB-doped polymer film is pumped, the light

![Fig. 1](image-url) Preparation and characterization diagram of the fiber source. (a) Specific steps for fabricating a random laser coupled with the optical fiber. (b and c) The optical photograph and micrograph of the fabricated fiber source. (d and e) The scanning electron microscopy (SEM) images of the polymer-coated fiber. The side view (d) and cross-sectional view (e) are shown. (f) The SEM image of TiO2 nanoparticles with an average diameter of around 50 nm.
with any directions is generated and scattered by the nanoparticles and the interface in the polymer layer. Random lasing resonance will be built up in the random system as the total gain is larger than the loss. Some of the obtained random lasers are attenuated by directly radiating from the gain layer into air and/or passing through the optical fiber surface into air, and some are coupled into the optical fiber for long-distance transmission and emission from the end surface of the fiber. According to the refraction law and the total reflection condition, the radiation angle $\gamma$ is greater than $39.91^\circ$ relative to the axis of the fiber when the random lasing is emitted from the two ports of the fiber core as shown in Fig. S2 (ESI†). Based on the above analysis, the random lasing considered as a uniform plane light source at the fiber port and has a circular spot in real space in Fig. 2d. When a random laser passes through a lens, the beam becomes ring-shaped corresponding to the momentum space as shown in Fig. 2e.

Fig. 3a presents the absorption and photoluminescence spectra of the DCJTB film with a concentration of 1.5 mg mL$^{-1}$, demonstrating strong absorption for 532 nm pumping pulses and a fluorescence peak at about 624 nm. Thus, the DCJTB molecules in the random system can be excited well by the pumping pulses and the emission spectra of the random laser at various pump power densities are shown in Fig. 3b. Only a broad spontaneous emission spectrum centered at 628 nm is observed when the pump power density is less than 0.1591 MW cm$^{-2}$. As the pump power density exceeds 0.1591 MW cm$^{-2}$, several discrete narrow peaks with a linewidth of about 0.6 nm are clearly observed, indicating that coherent resonant feedback is built up in the sample. To calculate the effective cavity length of the random laser, the power Fourier transforms (PFTs) of the spectrum at a power density of 0.9350 MW cm$^{-2}$ (Fig. 3b) are calculated and presented in Fig. 3c. The spatial dimensions can be obtained from the formula $p_m = \frac{m\lambda}{\pi}$, where $p_m$ is a Fourier component, $m$ is the order.
of the Fourier harmonic, $n$ represents the refraction index of the gain medium, and $L_c$ is the localized cavity dimension.\textsuperscript{38,39}

According to the statistical distribution of the cavity length in the illustration, the effective optical cavity length $L_c$ is calculated to be 122.53 $\mu$m ($n = 1.4$). The variations of peak intensity and full width at half maximum (FWHM) of the random lasing mode at 635 nm with the pump power density are shown in Fig. 3d. When the pump power density exceeds 0.1335 MW cm\textsuperscript{-2}, the peak intensity rapidly increases and the line width sharply decreases to sub-nanometer with the increasing pump power density. This significant feature indicates that the fabricated random laser has a working threshold of about 0.1335 MW cm\textsuperscript{-2}.$^\text{40,41}$

The effect of pumping angle $\varphi$ on the performance of random lasing is also carefully studied and shown in Fig. S3 (ESI\textsuperscript{†}). Several spikes are generally observed by changing $\varphi$ from 20° to 160° at a pump power density of 0.6565 MW cm\textsuperscript{-2} (Fig. S3b), demonstrating that the multi-mode random lasing can be achieved in an extreme wide angle range. The corresponding threshold remains almost unchanged when the pumping angle $\varphi$ increases from 20° to 160° as shown in Fig. S3c.$^\text{†}$ Specifically, the threshold remains at a small value of 0.1194 MW cm\textsuperscript{-2} when the range of $\varphi$ is [60°, 130°]. The stability of the threshold gives the random laser wider implementation conditions.

The effect of thickness $d$ of polymer coating on the output performance of fiber-coupled random lasers is also demonstrated as shown in Fig. S4 (ESI\textsuperscript{†}) by carefully controlling the dipping times. When the random lasers are vertically pumped by the pulses with a power density of 0.2188 MW cm\textsuperscript{-2}, the random lasing intensity increases with $d$ rising from 0.2 to 24 $\mu$m, following a red-shift behavior of the center wavelength as large as about 10 nm. The larger thickness of the film can provide larger gain for random lasing. A longer period in the gain region leads to a strong reabsorption of dye molecules due to the overlap between the absorption and emission spectra of the DCJTB film as shown in Fig. 3a and then induces the spectral red shift of random lasing.$^\text{38,42}$ The wavelength shift phenomenon provides an optional approach to regulate the emission wavelength of the random laser by controlling the thickness $d$ of the coated polymer. In addition, the intensity of random lasing after propagation at different distances in the optical fiber is detected and shown in Fig. S5 (ESI\textsuperscript{†}) by maintaining the same pump position and power density. Coherent random lasers can still be detected with propagation distance $L$ increasing to 20 cm, although the intensity decreases with the propagation distance. The result shows that the designed fiber source could be used in integral optics.

The emission directionality of the designed sample is demonstrated in Fig. 4b and S6 (ESI\textsuperscript{†}) by keeping the detection distance as 25 mm from the pumping position and changing the detection angle $\alpha$ relative to the fiber axis in Fig. 4a. The relationship between the random lasing intensity and $\alpha$ at a pump power density of 0.9947 MW cm\textsuperscript{-2} is shown in Fig. 4b. A maximal intensity is observed in a very small angular range.
along the fiber axis which is 33 times as much as that in the 45° direction. This proves that the proposed fiber source has good directionality, which is better than that of the plasmonic random laser on the optical fiber end facet. Moreover, the robustness of the sample is revealed and shown in Fig. 4c and d by recording the spectrum 100 times at a pump power density of 0.7758 MW cm$^{-2}$. Notably, the intensity of the random laser is relatively stable after 100 measurement cycles, indicating that the random laser has good robustness.

The color of random lasing from the fiber source can be flexibly switched by integrating different color random lasers in one optical fiber as shown in Fig. 5a. A triple-state-switch-
able random laser is fabricated as shown in Fig. S7a (ESI†) by using the gain material Pyrromethene567 (PM567) for the yellow and DCJTB for the red random laser. Thus, a flexible color-switchable random laser among yellow, red and yellow-red lasers can be achieved by changing the pump position. When the yellow random laser is pumped by 532 nm pulses (Fig. S7b, ESI†), the emission spectra are obtained shown in Fig. 5b at different power densities. Similar to the above-mentioned red random laser, the coherent yellow random laser is obtained with a FWHM of about 0.5 nm when the pump power density is larger than the threshold of 0.1193 MW cm\(^{-2}\) as shown in Fig. 5c. Moreover, a yellow-red dual-color random laser can be obtained when the two polymer random lasers are simultaneously pumped as shown in Fig. S8.† The results

![Diagram of the experimental setup](image)

**Fig. 6** Application of the random laser in speckle-free full-field imaging. (a) Schematic of the experimental setup for microscopic imaging. The random laser and He–Ne laser are separately used as the illumination sources. (b and c) Optical images of the speckle pattern by using the obtained random laser (b) and a 633 nm laser (c) as the illumination sources. The calculated speckle contrasts are shown in the left corner of each image. (d and e) Optical images of the lens paper under the illumination of the random laser (d) and a 633 nm laser (e). (f and g) Optical images of the lens cleaning paper under the illumination of the obtained random laser (f) and a 633 nm laser (g) in a strong scattering environment by introducing a frosted glass in the illumination path.
demonstrate the potential of the proposed fiber source in achieving a multi-color random laser and a color-switchable random laser.

The ability of the fabricated random laser as an illumination light source of the microscopic imaging in eliminating coherent artefacts is further demonstrated in Fig. 6. The optical setup of microscopic imaging is shown in Fig. 6a. The laser is collected by using a lens to illuminate the object and/or the scattering film to form the illumination path. An objective lens (100×) is used to collect the object information and a camera is used to record the resultant image for the imaging part. Firstly, the role of preventing speckle formation is directly revealed by the images in Fig. 6b and c when the random laser and a 633 nm He–Ne laser directly pass through a frosted glass with a coarse surface, respectively. The speckle is obviously observed in Fig. 6c when using the narrowband laser, but the obtained images with random lasing illumination show a uniform optical field without any observable speckle in Fig. 6b. To quantitatively analyze the speckle suppression effect, the speckle contrast \( C \) is defined as \( C = \frac{\sigma_{I}}{\langle I \rangle} \), where \( \sigma_{I} \) is the standard deviation of the image intensity and \( \langle I \rangle \) is the average intensity of the image.\(^{17,22}\) The calculated speckle contrasts are \( C = 0.41 \) for 633 nm laser illumination and \( C = 0.02 \) for random lasing illumination which is lower than that of the polymer fiber random laser.\(^{36}\) The extremely low speckle contrast is attributed to the low spatial coherence and the ring-shape in momentum space of our designed random lasing.\(^{30}\) Then, the ability of improving the image quality of the fabricated random laser is approved by comparing the images of lens paper with random structures illuminated by the two light sources described above. The image based on the narrow-band laser exhibits obvious speckle patterns within and near the paper fibers. These artificial intensity modulations corrupt the image as shown in Fig. 6e. However, the random laser can eliminate well the coherent artefacts and produce a clear image of the random object in Fig. 6d. Finally, the role of eliminating the speckle of random lasing is demonstrated in a strong scattering environment by introducing a scattering film of frosted glass in front of the lens paper in the illumination path. Under 633 nm laser illumination, high spatial coherence results in a strong speckle phenomenon and the image of the lens paper is completely submerged in speckle noise beyond recognition as shown in Fig. 6g. When illuminating with the obtained random laser, speckle noise is precluded due to the low spatial coherence, leading to a uniform background signal and a clean image of the lens paper in Fig. 6f even in a strong scattering environment. These results further prove that the proposed fiber source has a good application prospect in the field of full-field speckle-free imaging and display.

4. Conclusions

A ring-shaped fiber random laser in momentum space is fabricated by coating a polymer film on the surface of a commercial optical fiber. The sample possesses novel, unique and complement features in comparison with conventional random laser devices. Firstly, this is a new approach of fabricating fiber sources by coupling a random laser and a commercial optical fiber. The obtained fiber source has the advantages of directional emission, flexibility, easy integration, low spatial coherence and so on. Moreover, it is very convenient to switch the output color of the fiber random lasing by integrating different color random lasers on one optical fiber and mechanically controlling the pumping position. Secondly, the output spots of the random laser from the sample are circular in real space but ring-shaped in momentum space due to the unique coupling mechanism of the random laser and optical fiber. This unique feature provides a new way to shape the beam for further suppressing the coherent noise and achieving super-resolution imaging. Finally, the red and yellow random lasers coupled with the optical fiber have low thresholds of 0.1335 and 0.1193 MW cm\(^{-2}\), respectively. The low working threshold assures the good stability and repeatability of the sample. As a light source, the achieved random laser shows great potentiality in high-quality speckle-free microscopic imaging in different scattering environments. The newly designed fiber source may lead to further practical applications in the fields of sensing, \textit{in vivo} biological imaging, high brightness full-field illumination and interference detection.

Conflicts of interest

The authors declare no conflict of interest.

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Notes and references
