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Lasing characteristics of random cylindrical microcavity lasers

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Room-temperature lasing characteristics of random cylindrical microcavity lasers, which can be realized by coating a layer of random gain medium onto the surface of an optical fiber with various diameters, was studied experimentally. It is shown that closed-loop random modes excited inside the random gain medium are strongly confined along the radial direction so that the spacing of lasing modes is controlled by the diameter of cylindrical microcavity. In addition, lasing threshold of the random gain medium can be reduced by an order of magnitude under the influence of radial optical confinement. © 2011 American Institute of Physics. [doi:10.1063/1.3670501]

Coherent random lasing has been observed from strong scattering media with high optical gain such as highly disordered ZnO thin films and nanocomposite films with ZnO powders.^{1,2} Although narrow linewidth can be obtained from these random lasers via formation of closed-loop random cavities, high pumping threshold, and multiple lasing modes of random media hinder their practical applications.³ In order to improve the lasing characteristics of random lasers, efficient optical confinement of random media was used to suppress the excitation of random modes. For example, planar random microcavity lasers had been proposed to improve the emission characteristics of random media via one-dimensional optical confinement of Bragg reflectors.⁴ In this Letter, the use of cylindrical microcavity is proposed to control the modal characteristics of random lasers.^{5,6} This is possible as total internal reflections at either outer boundary or both inner and outer boundaries of the cylindrical microcavity can suppress random modes, which are not in resonant with the cavity. Hence, the disadvantages of random lasers such as high pumping threshold and multiple-mode emission can be reduced by the use of radial optical confinement.

In the experiment, 100 mg of ZnO powder (purchased from Chemical Regent China), which has irregular shapes of dimension varies between 50 and 400 nm, was mixed with 1 mL of silicone resin (TMT-9022 A)⁷ to form the random medium. The mixture was spin-coated onto a quartz substrate and then baked at around 100 °C for 1 h. As a result, a thin film of ZnO/SiO₂ composite with thickness s of $\sim 2 \mu\text{m}$ was formed on the quartz substrate (see the inset of Figure 1). Figure 1 plots the typical room-temperature lasing characteristics of the ZnO/SiO₂ composite thin film under optical excitation by a quadruplet Q-switched Nd:YAG laser (at 266 nm) at pulsed operation ($\sim 6 \text{ ns}$, 10 Hz). A pump stripe of width less than $30 \mu\text{m}$ was formed on the sample surface by focusing the laser beam via cylindrical lens. The laser emission was collected from the edge of the sample through an objective lens. It can be shown that the composite film exhibits random lasing action with pump threshold, I_{th} , at around 60 kW/cm^2 . In addition, the number of lasing modes increases from ~ 4 to ~ 15 for the excitation pump increases

from $\sim 1.0 \times I_{\text{th}}$ to $\sim 1.4 \times I_{\text{th}}$, respectively. The mode spacing $\Delta\lambda$, which is dependent on the excitation power, is varied from ~ 0.15 to $\sim 0.5 \text{ nm}$ over the emission spectrum. In addition, linewidth of the lasing modes, $\delta\lambda$, is less than 0.3 nm . Lasing modes were also observed from the top and bottom surfaces of the sample (not shown). The polarization of the lasing light is dominated by TE polarization and TM modes are strongly suppressed by the waveguiding properties of the thin film structure. Hence, it is verified that the ZnO/SiO₂ composite thin film supports coherent random lasing action.^{1,2}

Random cylindrical microcavity lasers can be fabricated by coating a layer of ZnO/SiO₂ composite thin film onto the surface of a tapered optical fiber (Coring single-mode fiber with original diameter of $125 \mu\text{m}$) with diameter varies from ~ 40 to $\sim 125 \mu\text{m}$. Inset of Figure 2 shows the schematic of the random cylindrical microcavity laser. The fiber was dipped vertically into the mixture of silica resin and ZnO powder and then baked at around 100 °C for 1 hour. A layer of ZnO/SiO₂ composite film with thickness of $s \sim 2 \mu\text{m}$ was then formed over the length of the optical fiber. Figure 2 plots the emission spectra versus pump intensity of the random cylindrical microcavity at room temperature. A pump

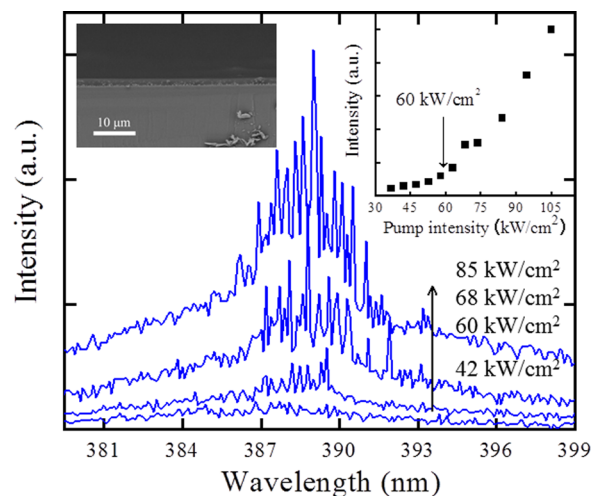


FIG. 1. (Color online) Room-temperature emission spectra of the ZnO/SiO₂ nanocomposite thin film versus pump intensity. An inset (left) shows the scan electron microscope image of the ZnO/SiO₂ nanocomposite thin film.

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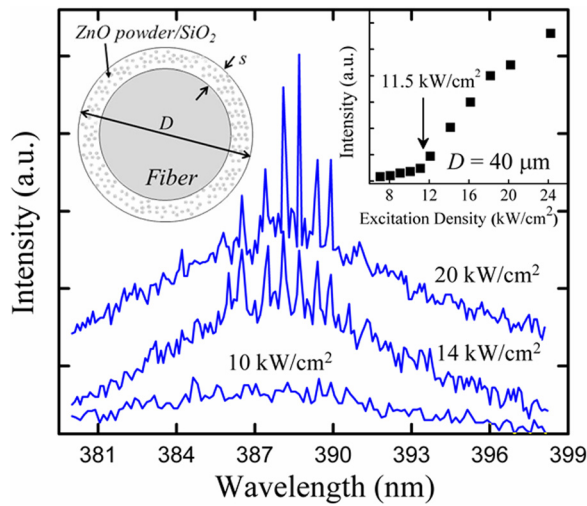


FIG. 2. (Color online) Room-temperature emission spectra of the random cylindrical microcavity laser (coated with ZnO/SiO₂ nanocomposite thin film) with $D = 40 \mu\text{m}$ versus pump intensity. The insets show the corresponding light-light curve (right) and schematic diagram of the random cylindrical microcavity laser (left).

stripe with width of less than $30 \mu\text{m}$ was focused on the surface perpendicular to the length of the cylindrical microcavity with diameter, D , equal to $\sim 40 \mu\text{m}$. Light was collected from the excited region of the cylindrical microcavity through an objective lens. The pump threshold, I_{th} , of the cylindrical microcavity is $\sim 11.5 \text{ kW/cm}^2$. It is noted that the number of lasing modes observed from the cylindrical microcavity is much less than that of the ZnO/SiO₂ composite thin film even the pump intensity is more than $1.7 \times I_{\text{th}}$. The mode spacing, $\Delta\lambda$, which is roughly equal to $0.7 \pm 0.1 \text{ nm}$, is less dependent on the pump intensity. The variation of $\Delta\lambda$ is due to the presence of random medium slightly alters the resonant conditions of cylindrical microcavity laser. Furthermore, TE modes are dominated in the lasing spectra as the TM modes have higher sensitivity to the surface roughness of the cylindrical microcavity.⁸

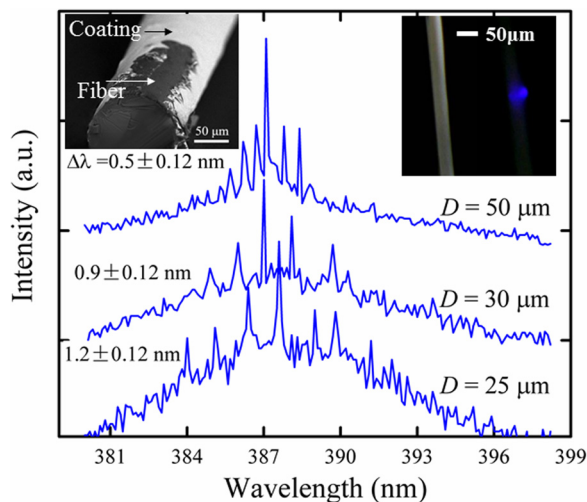


FIG. 3. (Color online) Emission spectra of the random cylindrical microcavity lasers (coated with ZnO/SiO₂ nanocomposite thin film) with $D = 25, 30$ and $50 \mu\text{m}$. The inset (right) shows the near field profile of the random cylindrical microcavity with $D = 50 \mu\text{m}$. A cross-sectional view of the fiber with $D \sim 125 \mu\text{m}$ is also shown in the inset (left).

Figure 3 plots the room-temperature emission spectra of the random cylindrical microcavity laser versus diameter D with the pump intensity maintained at around 1.5 to $1.7 \times I_{\text{th}}$. It is observed that the values of $\Delta\lambda$ increases with the decrease of D . This indicated that the presence of cylindrical microcavity suppresses the excitation of random modes and the resonant conditions of lasing modes are mostly dependent on the geometry of the cylindrical microcavity. Fast Fourier transform of the random lasing spectra given in Figure 1 has shown that the minimum closed-loop cavity length is about $10 \mu\text{m}$ (i.e., diameter of $\sim 3.2 \mu\text{m}$).⁹ This indicated that the thickness of ZnO/SiO₂ thin film is shorter than the scattering mean-free path of the random modes and hence the resonant cavity modes should be supported inside the random cylindrical microcavity. The inset of Figure 3 shows the near field profiles of the random cylindrical microcavity with $D = 50 \mu\text{m}$. It is observed that the lasing light was only emitted from the radial direction of the cylindrical microcavity.

As the scattering mean-free path is longer than the thickness of the ZnO/SiO₂ composite thin film,⁹ it is possible to assume that there may have three possible resonance modes to be supported by the random cylindrical microcavity (see the inset of Figure 4): (i) originates by total internal reflections of light only at the interface between air and ZnO/SiO₂ composite layer—whispering gallery modes (WGMs), (ii) total internal reflection of light at the interface between air and ZnO/SiO₂ (i.e., for $\theta > \theta_c$) and transmits partially at the interface between ZnO/SiO₂ composite layer and optical fiber (i.e., $\phi < \phi_c$)—WGMs of 2nd type,¹⁰ where θ_c and ϕ_c are the critical angles, and (iii) total internal reflection between the two interfaces (air and ZnO/SiO₂ composite as well as ZnO/SiO₂ composite and optical fiber)—waveguide modes in the cylindrical microcavity. As a result, random modes generated within the ZnO/SiO₂ thin film will be emitted to the surrounding through the coupling with one of these resonance conditions. This is the reason why the emission spectra of random medium are significantly modified by the presence of the cylindrical microcavity. The mode spacing

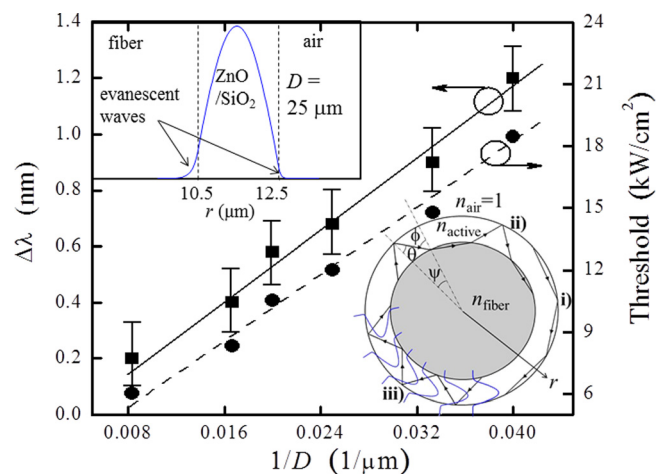


FIG. 4. (Color online) Plots of $\Delta\lambda$ and I_{th} versus $1/D$ of the random cylindrical microcavity laser at room temperature. An inset (top) shows the calculated radial distribution of the guided fundamental TE mode inside the cylindrical microcavity with D and s equal to 25 and $2 \mu\text{m}$, respectively. A schematic diagram, which describes the resonance mechanisms of a cylindrical microcavity laser, is also shown in the inset (bottom).

$\Delta\lambda$ due to (i), (ii), and (iii) can be approximated by using $\Delta\lambda \approx \lambda_o^2/\pi n_{\text{active}}D$, $\Delta\lambda \approx \lambda_o^2/L_c$, and $\Delta\lambda \approx \lambda_o^2/\pi n_{\text{eff}}D$, respectively, where n_{active} is the refractive index of the ZnO/SiO₂ composite thin film, L_c is the round-trip length (i.e., dependent on D and s) of the 2nd type WGMs and n_{eff} is the effective refractive index of the ring cavity.¹⁰ Therefore, the resonant mechanisms of the random cylindrical microcavity can be investigated by studying the variation of $\Delta\lambda$ versus $1/D$.

Figure 4 plots $\Delta\lambda$ and I_{th} of the random cylindrical microcavities with different values of D . It is noted that the variation of $\Delta\lambda$ is directly proportional to $1/D$, this indicated that the resonant conditions (i) and (iii) can be used to describe the dependence of $\Delta\lambda$ on D . The linear fitting of the numerical data of $\Delta\lambda$ versus $1/D$ has found that the corresponding slope is $\sim 3.125 \times 10^4 \text{ nm}^2$ so that the corresponding refractive index (either n_{active} or n_{eff}) can be ~ 1.534 at $\lambda_o \sim 388 \text{ nm}$. The dependence of L_c on D can also be studied if we assumed that the refractive indexes of optical fiber and the ZnO/SiO₂ thin film are equal to ~ 1.465 and ~ 1.534 , respectively.¹⁰ However, our calculation had found that the 2nd type WGMs is prohibited inside the cylindrical microcavity with these parameters taken into calculation. This is because the necessary conditions for $\theta > \theta_c (=40.7^\circ)$ and $\phi < \phi_c (=72.1^\circ)$ cannot fulfill simultaneously. In addition, periodic modulation of emission spectra, which is one of the spectral characteristics of 2nd type WGMs, was not observed in our experiment.⁹ Therefore, it is shown that resonant modes of (ii) was not supported by the proposed random cylindrical microcavity laser. In fact, the resonant mechanism of random cylindrical microcavity is likely to be (iii)—waveguide modes. First, the peak wavelength of the lasing spectra remains roughly unchanged with the presence of cylindrical microcavity. If resonant modes of (i) (i.e., WGMs) are supported, red-shift of lasing peak should be observed due to the increase of Q -factor. Q -factor of the random cylindrical microcavity laser with $D \sim 125 \mu\text{m}$ was found to be about $4 \times 10^3 (= \lambda_o/\delta\lambda)$, which is not as high as that of the WGMs.⁴ Second, due to the geometry of the cylindrical microcavity, evanescent waves are excited outside the interfaces of ZnO/SiO₂ layer. The inset of Figure 4 also plots the calculated radial distribution of the fundamental TE mode (i.e., higher order modes have lesser spatial overlap with the optical gain region so that they are ignored) for the cylindrical microcavity with $D = 25 \mu\text{m}$. Evanescent waves can be supported at

the interfaces of the ZnO/SiO₂ layer so that the proposed structure favors the excitation of guided modes. Lastly, it is noted from Figure 4 that the variation of I_{th} is inversely proportional to D . The linear dependence of I_{th} on $1/D$ has indicated that small cylindrical microcavity has high cavity loss. However, for microcavities support WGMs as described by (i), the variation of I_{th} should be less dependent on D .

The difficulty to support WGMs within the random cylindrical microcavity can be explained by their long propagation length. As the propagation length of WGMs is longer than that of the scattering mean-free path, a small change in propagation direction by scatterers will cease the formation of closed-loop resonant conditions. In contrast, waveguide modes are more stable inside the random medium as standing wave can be easily established within the two dielectric interfaces. In conclusion, a random cylindrical microcavity laser has been proposed and fabricated. We have verified that the resonant modes supported inside the random cylindrical microcavity is due to the waveguiding effect arisen from the relatively high refractive index of the ZnO/SiO₂ thin film when compared with that of the surrounding regions. The presence of cylindrical microcavities not only modifies the spectral characteristics of random media, the corresponding I_{th} can also be reduced from ~ 60 to $\sim 6 \text{ kW/cm}^2$ for the cylindrical microcavity with $D \sim 125 \mu\text{m}$.

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