Random lasers based on inverse photonic glass structure

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ABSTRACT

Random laser has attracted much attention because of its unique physical properties and potential applications in lighting, speckle-free imaging, biosensing, and photonic devices. In this work, we confirm that scattering plays a vital role in random lasing. Then, we investigate lasing properties of random film lasers with two scattering structures, including polystyrene microparticles and air voids embedded in a polymer matrix with organic dye serving as a gain medium. These two structures are called direct and inverse photonic glass, respectively. The result indicates that random lasers based on inverse photonic glass have a lower threshold. Following this achivement, we implemented inverse photonic glass into microspheres to obtain random microlasers of different sizes. Our work shows that inverse photonic glass structure is an excellent medium for random lasers with a wide range of sizes and dimensions. Especially, the obtained random microlasers are promising for applications in microsensors and photonic integrated circuits.

Keywords: Random laser; Inverse photonic glass; Microspheres.

1. INTRODUCTION

Random lasers have received significant interest due to their wide range of applications, including cost-effective light sources, speckle-free laser imaging, and rich physical properties [1]. The unique properties of a random laser are due to its structure. While a conventional laser uses an optical cavity to trap and amplify light, a random laser does not have a cavity. It relies on a disordered structure that amplifies light via multiple scattering [2, 3]. As a result, light scattering plays an essential role in the realization of a random laser. Up to date, various scattering structures have been used for random lasers, such as colloidal suspensions [4], semiconductor powders [4], nanowires [5], and animal and human tissues [6, 7].

The easiest way to fabricate a random laser is to embed scattering particles like the polystyrene (PS) microparticles into a polymer matrix with a gain medium. This structure can be called a direct photonic glass and has been demonstrated to be suitable for random lasers. Alternatively, a more advanced structure can be created from this direct photonic glass by chemically etching the PS particles. The obtained structure is highly porous and contains air voids in a polymer matrix, the so-called inverse photonic glass structure. Inverse photonic glass has recently been demonstrated as an excellent structure for random microlasers [8-10]. Since the refractive index contrast between the polymer and the air is generally higher than the PS particles and the polymer, the inverse photonic glass should offer a larger scattering strength which is more appropriate for random lasing. However, there is no report to confirm the advance of the inverse photonic glass over the corresponding direct photonic glass structure.

In this work, we experimentally demonstrate that a random laser based on an inverse photonic glass structure performs better, such as a lower lasing threshold than a random laser that relies on the corresponding direct structure.

2. EXPERIMENTAL

2.1. Fabrication of polymer films

Three types of polymer films have been fabricated: Film A is a plane film that does not have scattering particles or air voids; Film B and C have a direct and inverse photonic glass structure, respectively.

Firstly, 100 mg polyvinyl alcohol (PVA, from Sigma Aldrich) was dissolved in 2 mL water to form a PVA solution of 5 wt%. Secondly, 0.2 mL aqueous Rhodamine B (RhB, dye >95%, from Sigma Aldrich) solution (1 wt%) was added into the 2 mL PVA solution above and magnetic stirring at 80 °C for 15 minutes. Finally, RhB-PVA film was fabricated by using a micropipette to drop about 10 μL of the solution onto a glass substrate and then dry at 80 ^{o}C for 30 minutes (figure 1a, b).

To make a polymer film with a direct photonic structure, 0.5 mL aqueous suspension of monodisperse PS microparticles (10 wt%) was added to 1 mL RhB-PVA solution. Then, we dropped about 10 µL mixture onto a glass substrate and heated it at 80°C for 30 minutes (figure 1c, d). As a result, a dye-doped polymer film with embedded PS particles is obtained.

The fabrication process of an inverse photonic glass structure is the same as above, but additional chemical etching was carried out by immersing the film in ethyl acetate solvent for 48 hours (figure 1e). The solvent dissolves the PS particles but does not affect the host material. Thus a porous film is obtained [9].

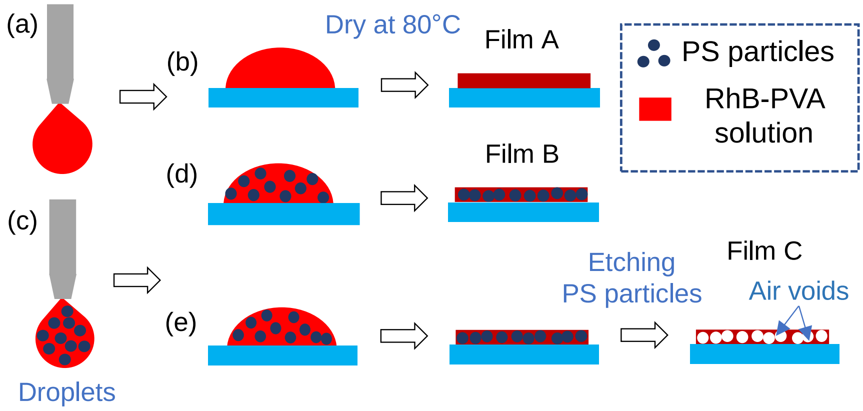

Figure 1. *Schematic diagram of the fabrication process of polymer films with various structures. (a,b) Fabrication of polymer plane film (film A). (c,d) Fabrication of PS film with direct photonic glass structure (film B). e) Fabrication of porous film with inverse photonic glass structure (film C).*

2.2. Fabrication of Microporous Spheres

Microporous spheres were fabricated using a self-assembled method and chemical etching process, as reported in [9]. Firstly, a droplet of RhB-PVA-PS mixture (prepared in section 2.1) was injected into a polydimethylsiloxane (PDMS) matrix (Sylgard 184 Silicon Elastomer from Dow Corning). Secondly, the droplet was dispersed into many smaller droplets. These droplets were then heated for 90 minutes to evaporate all water inside completely. After that, PDMS was removed by ethyl acetate solvent. Finally, PS particles were also etched with ethyl acetate, and porous polymer spheres with an inverse photonic glass structure were obtained.

2.3. Optical Measurement

We used a micro-photoluminescence (µ-PL) setup to study the dye-doped polymer films and microporous spheres. The pumping wavelength is 532 nm with a repetition rate of 10 Hz and a pulse duration of 8 ns (Canlas laser, CP 400-532). The focused laser beam spot is an ellipse with semi-major and semi-minor axes of 320 µm and 106 µm, respectively. The spot area is 0.106 mm^2. Emission from them was collected by a 10× objective and subsequently delivered to an AvaSpec-2048L (Avantes) for spectral recording. The spectral resolution is ~ 0.2 nm. Optical characterizations were carried out in the ambient air and at room temperature.

3. RESULTS AND DISCUSSION

3.1. Polymer film

Figure 2a shows a scanning electron microscopy (SEM) image (cross-section) of the RhB-PVA film with a thickness of about 10 µm. Under optical pumping, this film has strong photoluminescence and the emission spectra are plotted in figure 2b. It can be seen that the emission intensity with increasing pump fluence. However, the full width at half maximum (FWHM) of the spectra remains at about 50 nm, which is the characteristic of spontaneous emission. As expected, stimulated emission is not observed, suggesting there is not any optical feedback in the film. The result indicates that a scattering structure is needed to obtain stimulated and subsequently random lasing emission.

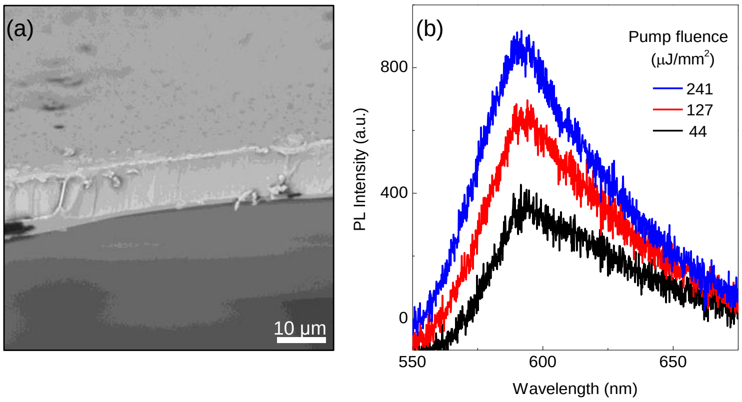

Figure 2. *a) SEM image of a dye-doped polymer film;*
b) Emission spectra of the film under different pump pulse fluences.

3.2. Polymer PS and porous films

Figures 3a and 3b present SEM images of a dye-doped polymer film with direct and inverse photonic glass structure, respectively. The PS particles and air voids in the polymer matrix can be observed clearly in the cross-section of the films. These structure scatters light strongly and should play a significant role in the realization of random lasing.

Upon optical pumping, these films can work as random laser sources. Figure 3c plots emission spectra of a dye-doped PS film as a function of pump fluences. It can be seen the evolution from fluorescent when the fluence is lower than 154 μJ/mm^2 to lasing emission when it reaches 269 μJ/mm^2. The evidence of lasing emission is the reduction of the spectral linewidth and the sharp increase of the PL emission. FWHM of the emission reduces from about 50 nm (at pump fluence around 80 μJ/mm^2 and lower) to around 7 nm at 269 μJ/mm^2. In addition, when the pump fluence increases 1.7 times (from 154 to 269 μJ/mm^2), the emission intensity increases more rapidly, 4 times (from 1200 to 4200 a. u.). This nonlinear dependence exhibits the threshold behavior of the random laser. Similarly, lasing emission is also obtained from the film with an inverse photonic glass structure. It is noticeable that the lasing spectrum is relatively smooth, and no spikers were observed. It is because the transport mean free path of this structure is around 3–7 µm which is much larger than the lasing wavelength (about 600 nm) [8]. In other words, the laser works in a diffusive regime with incoherent feedback.

Figure 3d plots the emission peak intensity versus pump fluences. It can be seen that the emission peak intensity increases linearly with the pump fluence until a certain threshold value. The lasing threshold of the film with a direct photonic glass structure is about 169 $\mu\mathrm{J/mm}^2$, which is higher than the threshold of ~108 $\mu J/mm^2$ of the film with the inverse photonic glass structure. It is understandable because the porous film has a higher refractive index contrast than the PS film. Indeed, refractive index contrast of these films are $\Delta n_i = n_{\mathrm{polymer}} - n_{\mathrm{air}} = 0.48$ and $\Delta n_d = n_{\mathrm{ps}} - n_{\mathrm{polymer}} = 0.12$ ($n_{\mathrm{ps}} = 1.6$ is the refractive index of the PS [8], $n_{\mathrm{polymer}} =1.48$ is the refractive index of polymer matrix [11]). As a result, the scattering mean free path of the porous film would be smaller than that of PS film. Thus the porous film laser has a lower lasing threshold [12, 13]. The result indicates the advantage of inverse photonic glass structure compared to the direct photonic glass structure.

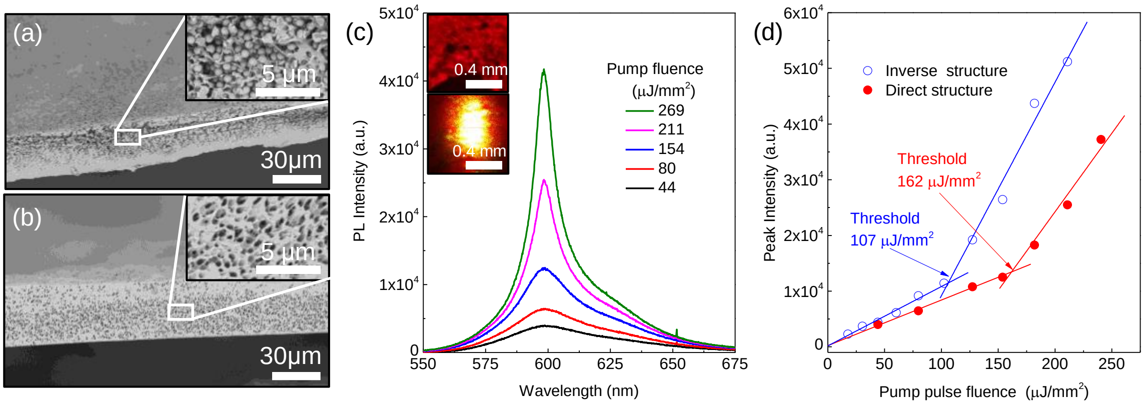

Figure 3. *a) and b) SEM and high-magnification SEM images of PS and porous polymer film, respectively. c) Emission spectra of the PS film under various pump fluences. d) Peak intensity of the PL emission of the PS film (red circle) and porous film (circle with blue outline) versus pump pulse fluences.*

3.3. Microporous Spheres

Figure 4 shows optical and SEM images of the fabricated microporous spheres. Their sizes are distributed in the range from 10 to 100 μm (figure 4a). Their spherical shape is well illustrated in figure 4b, while air voids and the polymer matrix are observed clearly in figure 4c.

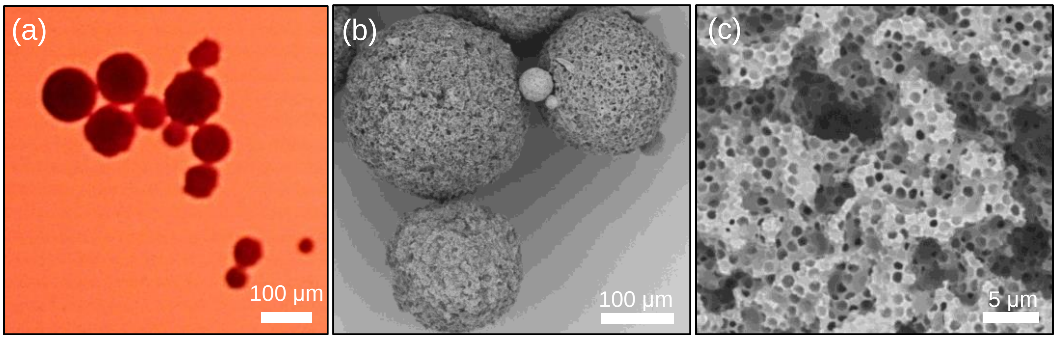

Figure 4. *a) Optical microscope image of fabricated microporous spheres with various diameters ranging from 10 to 100 μm. b) SEM image of typical microporous spheres. c) High-magnification SEM image of the porous structure.*

Figures 5a and 5b demonstrate the evolution from fluorescent to lasing of two porous spheres. Like the film random laser, when the pump fluence is smaller than the lasing threshold, the sphere emits spontaneous emission characterized by low intensity and a broad spectrum. When the pump fluence is greater than the lasing threshold, the emission intensity increases sharply, and the spectral linewidth of the emission becomes much narrower. The lasing wavelengths at the peak intensity of the 50 µm and 140 µm microspheres are 584.3 nm and 587.4 nm, respectively. That means the lasing wavelength of the larger sphere is 3.1 nm red-shifted compared with the smaller sphere. This phenomenon has been observed previously and explained by the reabsorption of the dye molecules [9]. Light in the larger sphere travels a longer path and the possibility of being obsorbed is higher. In addition, the shorter wavelength is absorbed more than the longer wavelength. Therefore, laser emission is red-shifted with increasing size.

The lasing threshold of the 50 µm microporous sphere is 37 µJ/mm^2, while it is 30 µJ/mm^2 for the 140 µm microsphere (figure 5c). That means the smaller sphere has a higher threshold than the larger sphere. It is because the light emission in the larger sphere can travel a longer path thus light can be amplified better compared with the smaller sphere.

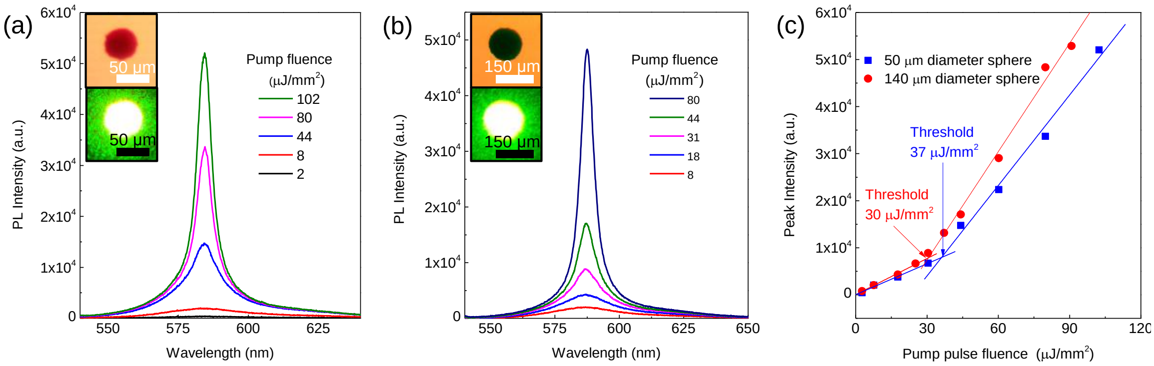

Figure 5. *a) and b) Emission spectra of a 50 µm and 140 µm diameter sphere under various pump pulse fluences, respectively. c) Corresponding PL peak intensity of these two spheres versus pump fluence.*

4. CONCLUSIONS

We have demonstrated that scattering plays a significant role in random lasing. Then, we fabricated polymer film with two scattering structures, the direct photonic glass structure (PS microparticles embedded in a polymer matrix) and the inverse photonic glass structure (air voids in a polymer matrix). The result indicates that a random laser based on the inverse photonic glass structure performs better, such as a lower lasing threshold. Owning the advantages of the inverse photonic glass structure, we implemented it into porous microspheres and the obtained microspheres can work as random microlasers. Our work provides a unique and simple technique for fabricating film-shaped and spherical random lasers with good characteristics.

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TÓM TẮT

Laser ngẫu nhiên dựa trên cấu trúc thủy tinh quang tử ngược

Laser ngẫu nhiên đang thu hút được sự quan tâm nghiên cứu vì chúng có các tính chất vật lý độc đáo và tiềm năng ứng dụng trong các lĩnh vực như chiếu sáng, tạo ảnh, cảm biến sinh học và thiết bị quang tử. Trong bài báo này, trước tiên chúng tôi chứng minh rằng tán xạ đóng vai trò quyết định trong việc tạo ra laser ngẫu nhiên. Sau đó, chúng tôi nghiên cứu các đặc tính phát quang của laser ngẫu nhiên từ màng polyme pha hoạt chất màu hữu cơ (đóng vai trò như môi trường khuếch đại) với hai đặc tính khác nhau: các vi hạt nhựa sắp xếp chặt chẽ và cấu trúc xốp với nhiều lỗ rỗng. Hai cấu trúc này lần lượt được gọi là thủy tinh quang tử thuận và ngược. Kết quả chỉ ra rằng laser ngẫu nhiên dựa trên cấu trúc thủy tinh quang tử ngược có ngưỡng phát thấp hơn. Cuối cùng, chúng tôi đã chế tạo thành công các hạt vi cầu có cấu trúc thủy tinh quang tử ngược và nghiên cứu được đặc trưng phát laser ngẫu nhiên của chúng. Dữ liệu thu được cho thấy thủy tinh quang tử ngược là cấu trúc tốt để tạo ra các nguồn vi laser ngẫu nhiên với kích thước và hình dạng khác nhau. Các vi laser ngẫu nhiên đã chế tạo được có triển vọng ứng dụng trong vi cảm biến và vi mạch tích hợp quang tử.

Từ khóa: Laser ngẫu nhiên; Thủy tinh quang tử ngược; Vi cầu.