Pulse-duration-dependent and temperature-tunable random lasing in a weakly scattering structure formed by speckles

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It is reported experimentally that low-threshold random lasing in a weakly scattering disordered structure formed by speckles using holography is achieved. It shows that the emission property of the structure is related directly to the pulse duration of the pump laser. If pumped by picosecond pulses, several spikes are observed in the emission. If pumped by a nanosecond laser, only a single dominant peak appears. The wavelength tunability of the random laser with a change in temperature has been demonstrated. The corresponding physical analyses have also been provided briefly.

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I. INTRODUCTION

Since the pioneering work of Ambartsumyan *et al.* [1], random lasers have been the subject of intense theoretical and experimental studies [2–10]. In particular, the discovery of intriguingly narrow spectral emission peaks (spikes) in certain random lasers by Cao *et al.* [4] has given an enormous boost to such a subject. The random laser represents a nonconventional laser whose feedback is mediated by random fluctuations of the dielectric constant in space. In general, there are two kinds of feedback for random lasing, intensity feedback and field feedback [9,10]. The former is phase insensitive, which is called incoherent or nonresonant feedback. The latter is phase sensitive, which can be regarded as coherent or resonant feedback.

The two types of random laser action cannot only be found in some strong scattering systems with gain [9–12], they can also be observed in active random media in weakly scattering regimes far from Anderson localization. For example, some authors have reported laserlike emission from several weakly scattering samples including conjugated polymer films, semiconductor powders, and dye-infiltrated opals [13–17]. In the strongly scattering regime, lasing modes have a nearly one-to-one correspondence with the localized modes of the passive system [9–11,18]. In contrast, the nature of lasing modes in weakly scattering open random systems is still under discussion, although several mechanisms have been proposed to explain such a phenomenon [19–22].

II. EXPERIMENTS

Schwartz *et al.* reported a work in which a speckle beam was used to control the disorder level of triangular structure [23]. Here, we report an experimental observation of a random laser in a regime of weak scattering with a disorder structure formed by speckles. The setup geometry is shown in Fig. 1(a) schematically. A ground glass was used to generate spatial speckle structure. A reference beam was introduced to implement interference for recording the distribution of the spatial speckle in the holographic recording material. Different distributions of the spatial speckle can be recorded by changing the place of the ground glass or the reference beam. The recording material used was dichromated gelatin (DCG) coated on optical glass. The thickness of the DCG used was 36 μm. After exposure, the DCG plate was developed in running water for 120 min at 20°C for developing sufficiently and to remove any residual dichromate, and then was soaked in a Rhodamine 6G solution with a concentration of 0.125 mg/ml of water at the same temperature for 60 min; a bath enabling the dye molecules to diffuse deep into the emulsion of the gelatin. Then, the DCG plate was dehydrated in turn by soaking it in 50%, 75%, and 100% isopropyl alcohol containing the Rhodamine 6G dye with the same concentration at 40°C for 15 min.

After dehydration, the DCG plate was baked at 100°C for 60 min in an oven. Figures 1(b) and 1(c) show the microscopic image of the disordered structure formed by the speckle, and it is a completely disordered structure. The average size of the speckle spot is about 1 μm. The concentration of the speckle spot is estimated to be on the order of magnitude of 10^9 per cm^3. The DCG is a phase-type holographic recording material with the refractive index of 1.52 and the modulation of the refractive index less than 0.1. From the transmission measurements, we can estimate the mean-free path $l^* > 80$ μm in the spectral range between 550 and 600 nm, which is much larger than the thickness of the system. This means that scattering in the structure is quite weak.

III. EXPERIMENTAL RESULTS AND DISCUSSIONS

We now turn to the investigations on the photoluminescence (PL) of the preceding dye-doped random samples under a pump field. Figure 2(a) shows the setup geometry for the measuring. A nanosecond- (ns-)pulsed Nd:YAG laser running at 532 nm with a repetition rate of 10 Hz, maximum pulse energy of 1500 mJ, pulse width of 8 ns, and a picosecond- (ps-)pulsed Nd:YAG laser running at 532 nm with a repetition rate of 10 Hz, maximum pulse energy of 40 mJ, and pulse width of 30 ps were used as the pumping sources, respectively. The beam diameters of the ns laser and the ps laser were 13 and 3 mm, respectively. The laser beam was incident on the surface of the optical glass substrate, then penetrated the substrate and

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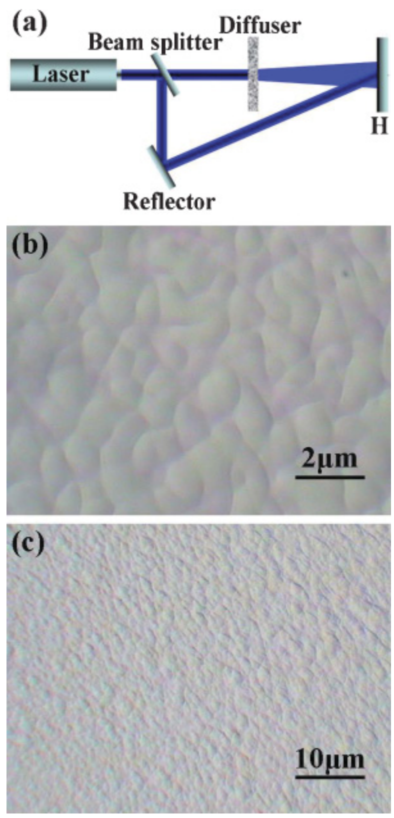

FIG. 1. (Color online) (a) Setup geometry of the disorder structure formed by speckles. H is the holographic recording material. (b) and (c) are the microscopic images of the disordered structure formed by speckles.

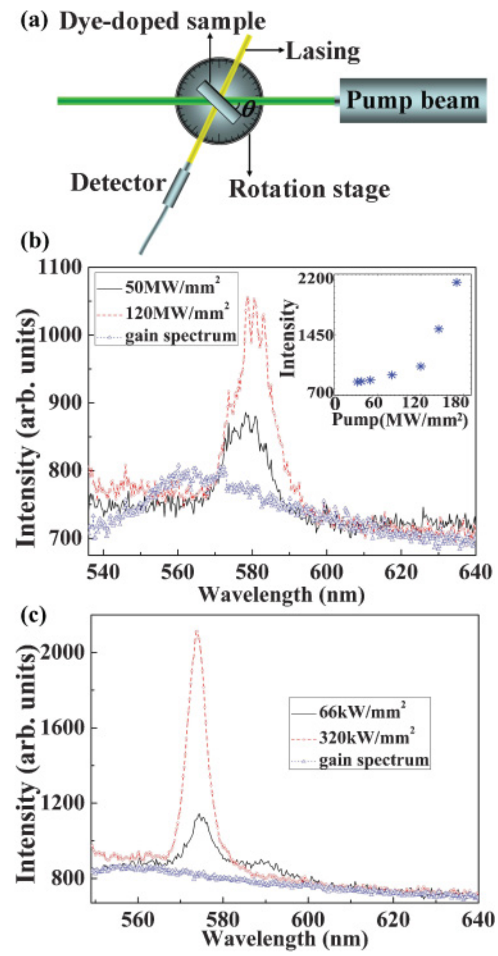

FIG. 2. (Color online) (a) Setup geometry for measuring random lasing. θ is the angle of the pump beam with respect to the surface of the substrate; it represents the orientation of the sample. (b) Emission pumped by the ps laser, and the inset shows the threshold is about 50 MW/mm^2 at $\theta = 25^\circ$. (c) Emission pumped by the ns laser at $\theta = 30^\circ$. In (b) and (c), the blue triangles are the gain spectrum of Rhodamine 6G excited by a 532-nm laser beam. The black solid line and the red dashed line represent the emissions with pumped energy around and much higher than threshold,respectively.

entered the dye-doped DCG medium. The detector was put close to the sample to collect as much energy as possible. The formed speckle structure is a quasi-two-dimensional diffusive system [24]. By rotating the rotation stage, the orientation of the speckle structure can be changed with respect to the laser beam. It results in the variation of the wavelength and the peak number of lasing.

The measured results for the emission spectra under the pump by the ps laser are plotted in Fig. 2(b). The blue triangles correspond to the PL spectra of Rhodamine 6G, and the red solid line represents the emission of the dye-doped sample at $\theta = 25^\circ$ under the pumping intensity of 120 MW/mm^2. The threshold of the random laser is about 50 MW/mm^2 as shown in the insets. It can be seen that many lasing modes are excited in the gain spectrum of Rhodamine 6G. The widths of these modes are found to be less than 0.4 nm, which looks like the emission of a random laser with coherent feedback [22]. The phenomenon is very similar to the previous investigations on the random laser in other weak scattering systems [13–17]. The origin of the phenomenon can also be understood by the previous theory for such a problem [19–22].

However, when the dye-doped random sample is pumped by the ns laser, a different phenomenon appears. Figure 2(c) displays the measured results when the pump energy is much higher (320 kW/mm^2) than and around (66 kW/mm^2) the threshold (the measured data is about 40 kW/mm^2). A single dominant peak is observed, and the profile of the peak is fitted by a Gaussian function and a Lorentzian function, respectively. It can be seen that the measured result is more coincided to the Gaussian profile [as shown in Fig. 3(a)]. The possible reason is that the process of enormous random scattering by speckle spots with different size, distance, and shape should be expressed by a Gaussian function (central limit theorem). When $\theta = 30^\circ$, the emission spectra at different pump energies are shown in Fig. 3(b). There is only a single dominant peak at 574 nm. The threshold value is found to be about 40 kW/mm^2, which is much lower than that pumped by the ps laser (50 MW/mm^2). The lasing becomes more obvious when the pump energy is higher than 66 kW/mm^2.

In fact, our sample is in the shape of a thin film; it cannot be considered as a strict isotropic structure. So, the property of the peaks also depends on the pump direction of the laser beam. In some directions, the case with two peaks can also be found. Figure 4(a) shows the measured results for the emission spectra of the disordered structure at different pump energies using the ns laser. There are two peaks of the emission located at 570 and 590 nm, respectively, the latter is much stronger

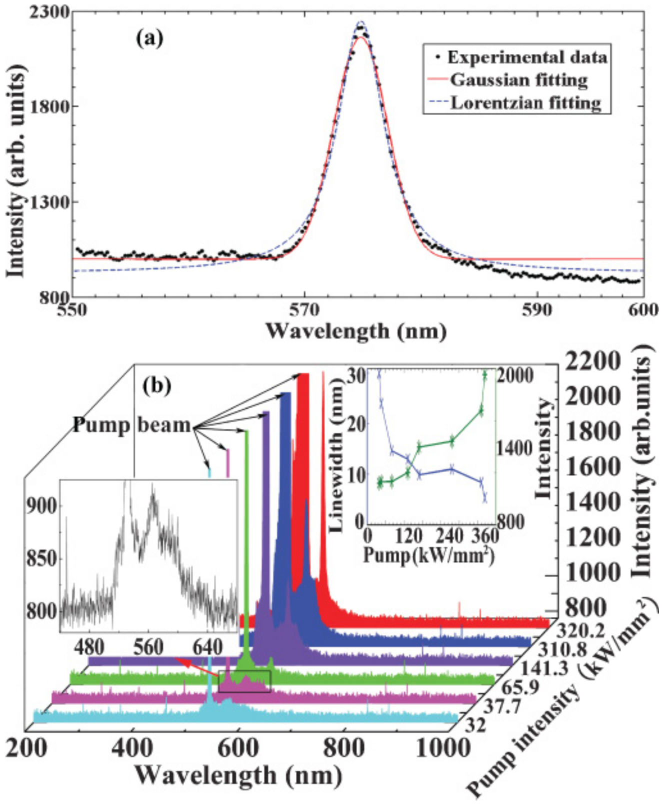

FIG. 3. (Color online) (a) Gaussian and Lorentzian functions fitted emission peak. (b) Measured emission spectra at different pump energies in the case of $\theta = 30^{\circ}$, and the inset shows the threshold is about 37.7 kW/mm^2. The left high spikes (marked by black arrows) correspond to a 532-nm pump laser beam, and the apparent width does not show its actual linewidth of the pump laser because of the saturation effect of the charge-coupled device (CCD) detector used. In the right inset of (b), the signal $\times$ represents the data of linewidth, the signal * represents the data of intensity.

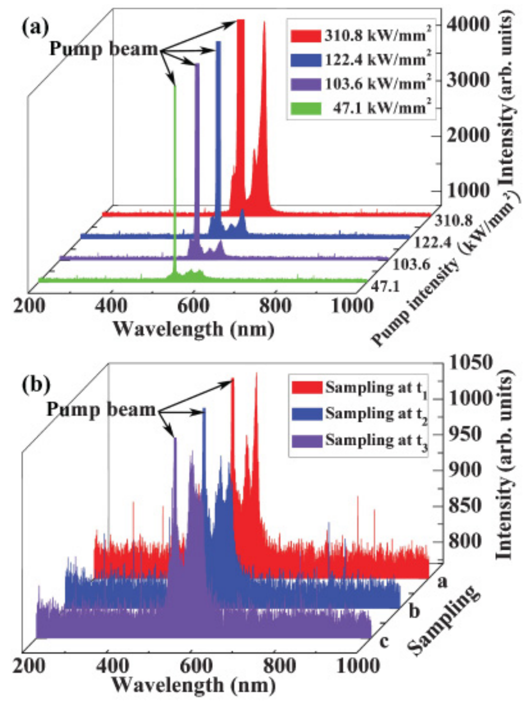

FIG. 4. (Color online) (a) Measured radiation spectra at different pump energies in the case of $\theta = 18^{\circ}$. (b) Competition of two peaks around the threshold value in the case of $\theta = 18^{\circ}$. The left high spikes (marked by black arrows) correspond to a 532-nm pump laser beam, and the apparent width does not show its actual linewidth of the pump laser because of the saturation effect of the CCD detector used. In (a) and (b), the legends from top to bottom represent the spectra from back to front in the figures.

than the former, and the latter becomes dominant when the pump energy is strong enough. When the pump energy is around the threshold value, the two peaks are in competition, which is similar to mode competition in a conventional laser see Fig. 4(b)]. In Figs. 4(a) and 4(b), the legends from top to bottom represent the spectra from back to front.

In addition, we find that the widths of the peaks change from 30 to 5 nm (see the right inset in Fig. 3(b)] when energy of the ns pump beam increases, which displays a character similar to intensity feedback [22]. In general, the intensity feedback corresponds to diffusion motion of photons in active random medium, when the photon mean-free path is much smaller than the dimension of the scattering medium but much longer than the optical wavelength [9,10]. It can be described theoretically by the diffusion equation for the photon energy density in the presence of a uniform and linear gain [1,9,10]. Obviously, it is not the case for the weak scattering system in the present paper because the mean-free path is much larger than the thickness of the sample. Our experimental results demonstrate that the phenomena similar to incoherent feedback can also be realized in our structure by choosing suitable pump sources. Besides, in our experiments, the phenomena similar to coherent feedback was never observed regardless of the high or low energy of the ns pump laser. In contrast, the phenomena similar to incoherent feedback was never observed regardless of the high or low energy of the ps pump laser. In previous investigations, one has observed the transition between coherent feedback and incoherent feedback by varying the amount of scattering in the gain medium [9,25]. In fact, our present paper illustrates that a similar transition can also be realized in the same random sample by changing the pump duration.

The phenomena can be understood in terms of the following analysis. For the case of ns laser pumping, the light will go through a path of about several meters in the period of one pulse. It means that the light will have much more time for scattering (i.e., has much longer effective interaction length). Therefore, the mode competition has finished, and one peak becomes dominant. That is the reason only a single peak is dominant in the emission as long as the pumping energy is higher. However, for the case of ps laser pumping, the light will go through a path of about several millimeters. It means that the effective interaction length is much shorter, so the mode competition cannot be finished even though the pumping energy is rather high. Besides, for the case pumped by the ps laser, the effective interaction length is very short, that is, the scattering times are much less. Therefore, much higher pump energy is needed for building up the lasing; it results in the high threshold.

Another interesting phenomenon for the present system is that the emission wavelength for the random laser can

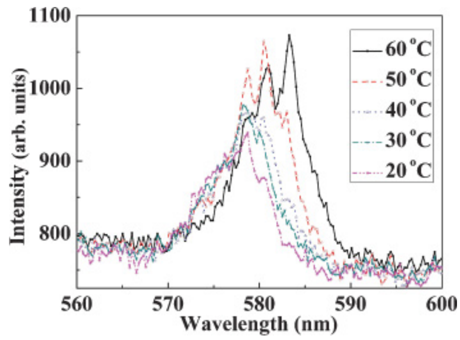

FIG. 5. (Color online) Tunability of wavelength vs temperature of the random laser.

be tuned through changing temperature. Figure 5 shows the measured spectra of lasing emission at different temperatures. It can clearly be seen that the emission peaks shift toward the long wavelength with the increase in temperature. The lasing will be kept so long as the frequency of the emission is still inside the gain profile when the temperature changes. In fact, the temperature tuning for the random laser by infiltrating sintered glass with laser dye dissolved in a liquid crystal had been investigated in a previous work [6]. The diffusive feedback was controlled through a change of refractive index of the liquid crystal with temperature. However, such a tuning was employed to turn random lasers on and off. In contrast, the wavelength tuning can be realized in the present system. The phenomenon originates from the special property of the material. With the changes in temperature, the light path changes due to variation of the distance between the two adjacent speckle spots. It makes the variation in the effective length of the interaction, and then the lasing wavelength is changed.

Finally, it should be pointed out that, the DCG can be coated on a soft substrate, such as polyethylene terephthalate (PET). So, the speckle structure recorded by holography can be formed as a soft film. On the other hand, the holography recorded speckle structure is pretty stable. Hence, the output of this kind of laser has good stability and repeatability. It will be beneficial for actual applications. We did implement the speckle random laser using DCG coated on the PET soft substrate, and the same results mentioned previously were obtained.

IV. SUMMARY

To summarize, we have fabricated a weakly scattering disordered structure formed by speckles using holography. In such a system with gain, we have observed low-threshold random lasing with two kinds of feedback, incoherent and coherent, for the same sample in the case pumped by ns and ps lasers, respectively. The wavelength tunability of the random laser with a change in temperature has been demonstrated. Our results cannot only get a deeper understanding on the open question about the properties of the lasing modes in a weakly scattering regime, they can also benefit by some applications, such as remote temperature sensing in hostile environments.

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