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Random lasing with scatterers of diameters 20 nm in an active medium

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Abstract

In this letter we reported random lasing achieved in MEH-PPV/glass waveguide with the 20-nm TiO_2 scatterers in diameter significantly reduced in size compared to submicrometer of TiO_2 scatterers in films or suspensions previously reported on random lasing. The spectral lines were dramatically narrowed by almost two orders of magnitude compared with those excited by Xenon lamp. The amplified spontaneous emission (ASE) was identified as the dominant mechanism in our system. Thouless number [H. Cao, Wave Random Media 13 (2003) R1] should be $\delta \equiv \delta\nu/(\mathrm{d}\nu/\mathrm{d}N) = 1.6 \times 10^6 \gg 1$. It indicates again that there is no coherent amplification with coherent feedback due to Anderson localization of light in our case.

In recent years significant advantages of random lasers such as low loss and cost, high gain, and even thresholdless feature have remarkably promoted the development of random lasers for potential applications in photonics [1,2]. Conventional lasers require a precise design to configure the cavity for lasing and light scattering is highly depressed for laser action. In 1968, Letokhov calculated the amplification and scattering of a random medium with gain using the diffusion equation and pointed out that laser action is possible in these media due to the process of diffusive feedback [3] and lasing does not rely on the coherent feedback provided by resonant cavities. This type of laser action has been known as "random laser" [4].

The idea for random laser originally proposed by Letokhov was demonstrated by the experimental discovery in Lawandy's group in 1994 [5]. The introduction of suspending TiO_2 into the Rhodamine dye solution was found to cause a remarkably spectral narrowing in the emission spectrum of

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Rhodamine dye when the pump light intensity exceeded the threshold. Pioneering work of random lasing in TiO_2 nanoparticles doped in MEH-PPV/polystyrene film was done by Hide et al. [6] in 1996.

To our knowledge, the diameters of scatterers in submicrometer were so far reported for experimental investigation on random lasing. In this letter we report, for the first time, the experimental observation of random lasing using the 20-nm TiO_2 scatterers in poly(2-methoxy-5-(2′-ethylhexyloxy)-1,4-phenylene vinylene) (MEH-PPV)/glass waveguide.

MEH-PPV was dissolved in tetrahydrofuran (THF) solution. Four films of (1:0), (1:1), (1:2), and (1:3) were prepared using dip-coating and they contained MEH-PPV 1 mg/mℓ for (1:0), [MEH-PPV(1 mg) + TiO_2(1 mg)]/mℓ for (1:1), [MEH-PPV(1 mg) + TiO_2(2 mg)]/mℓ for (1:2), and [MEH-PPV(1 mg) + TiO_2(3 mg)]/mℓ for (1:3), respectively. Fig. 1 shows the emission spectra excited by Xenon lamp with F-4500 fluorescence spectrometer. The full widths at half maximum (FWHM) of spectra for films (1:0) and (1:1) are 124 and 103 nm, respectively.

A Spectra-Physics Ti:Sapphire laser delivering 3 μJ, 150 fs pulses at 544 nm with a repetition rate of 1 kHz was used as a excitation source. A schematic diagram for the experimental configuration is sketched in Fig. 2. An adjustable slit and a cylindrical lens were used to couple the beam into a stripe with a width of 300 μm and a variable length. Lasing action from the current stripe design (one-dimensional gain region) was more easily analyzed than the lasing action from a two-dimensional disk. Waveguide structure was found to show only one transverse electric (TE) mode without transverse magnetic (TM) modes. Most excitations were stimulated to emit into the waveguide modes, and thus a larger fraction of the light came out of the edge.

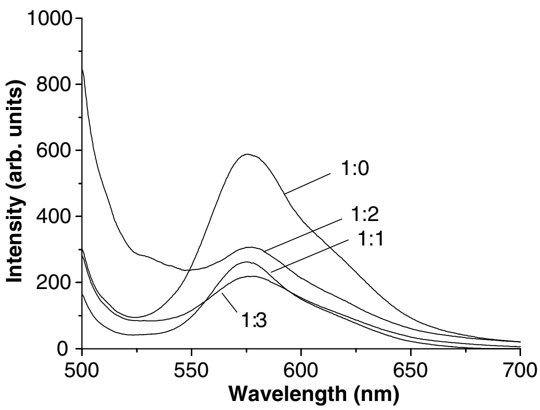

Fig. 1. Emission spectra of samples excited by Xenon lamp.

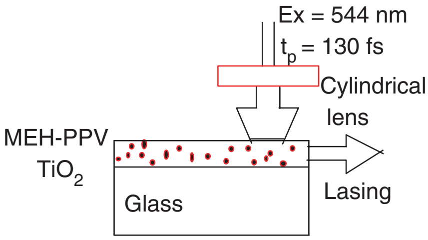

Fig. 2. Schematic of experimental configuration.

Morphology-dependent resonance (MDR) [7] takes place while the conditions of $d \geqslant \lambda$ and $n_{sca} > n_{sur}$ are satisfied, where d and λ denote the diameter of random scatterers and the excitation wavelength, and n_{sca} and n_{sur} represent the refractive index of the scatterers and the surrounding medium. Multi-scattering, possibly associated with refractive index and size of the scatterers, input and output directions of light and propagation inside the scatterers, and so on, makes the system lasing more complicated. Suspended scatterers are unavoidable in spite of ultrasonic vibrator used for the deposition of the submicrometer scatterers in some solutions. For this reason, the 20-nm TiO_2 scatterers in diameter were used for the preparation of high quality waveguide adopted in the current experiment. Oliveira and Lawandy [7] came to a conclusion that the scatterers are too small to maintain resonance with high Q-factor, our results, however, demonstrate that lasing actions in MEH-PPV/TiO_2 (1:1) doped film are much stronger than that in neat MEH-PPV (1:0) film (Fig. 3) using the same experimental condition as [7]. The FWHMs of samples (1:0) and (1:1) are 9 and 7 nm, respectively. ASE occurs even when the gain coefficient is small since the spontaneously emitted photons are waveguided to travel a longer distance through the gain medium, where they are amplified by stimulated emission. ASE appears at

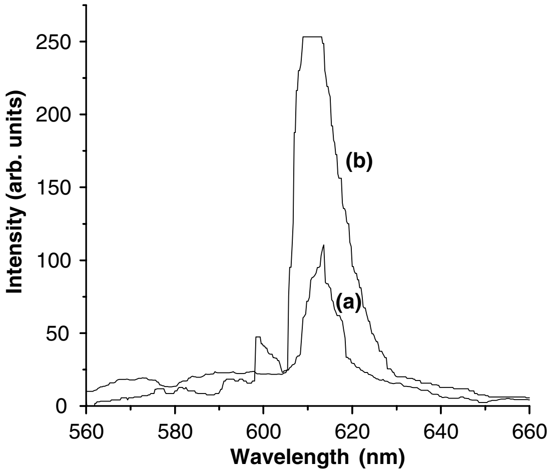

Fig. 3. Emission spectra: (a) MEH-PPV (1:0) film; (b) MEH-PPV/TiO_2 (1:1) film.

the wavelength peaking at the spontaneous emission spectrum where the gain is highest. The ASE wavelength is shifted to the red region at 610 nm in the emission spectra due to the near band edge absorption which reduces the net gain [8].

Fig. 4 shows the excitation dependence of FWHMs and the integrated intensities emitted from the edge. As excitation intensity is elevated, FWHMs of spectral lines decrease, whereas the spectral integrated intensity of the edge emitting increases.

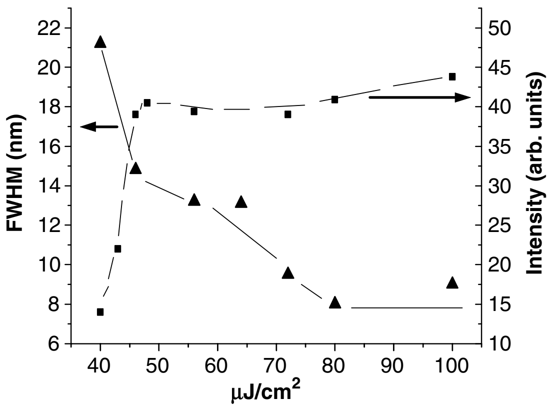

Fig. 4. The dependence of FWHMs of spectra (left y-axis) and the spectrum integrated intensities emitted from the edge (right y-axis) on the exciting power μJ/cm^2 of a pulse.

The spectral narrowing mechanisms are still in debate and a number of mechanisms leading to spectral narrowing were proposed as a result of cooperative superfluorenscence [9], bi-excitonic emission [10] and collective ASE [11]. Most of the reports pointed out that ASE is the main mechanism leading to the spectral narrowing. To determine the mechanism of lasing in our system two aspects in ASE were analyzed as the follows. The first is that the spectral lines were broadened at a short length of stripe and the spectral width was found narrowed with increasing the length of excitation stripe (Fig. 5). Various lengths of the excitation stripe were tested for $\ell = 1.06, 0.85, 0.76, 0.68, 0.59, 0.51, 0.49, 0.46$ and 0.42 mm, corresponding to FWHMs from 7 to 21 nm for sample (1:1). FWHM = 21 nm was found still narrower than FWHM = 103 nm for $\ell = 0.42$ mm for the same sample using a Xenon lamp excitation as plotted in Fig. 2. It implies that the excitation exceeds the intensity threshold. A double-peak emission was also found to appear in the spectra at intermediate pumping levels. Examination of Fig. 5 indicated that lasing modes changed with the pumping stripe length.

Secondly, in the regime of ASE the output intensity from one end of the stripe should be given by [8]

$$I(\lambda) = \frac{A(\lambda)I_p}{g(\lambda)}\{\exp[g(\lambda)\ell] - 1\}. \quad (1)$$

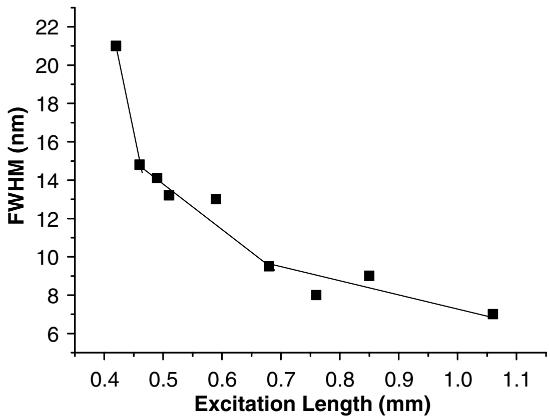

Fig. 5. Emission spectra of sample MEH-PPV/TiO_2 (1:1) film under the excitations with the various stripe lengths.

where A is the constant related to the cross-section for spontaneous emission, I_p is the pump intensity, g is the net gain coefficient or the gain minus the loss, and ℓ is the length of the pumped stripe. Fig. 6 illustrates that the dependence of the emission intensity on the excitation stripe length. The solid line fits to the experimental data using Eq. (1). If superfluorescence or biexcitonic emission is the mechanism responsible for spectral narrowing, the emission spectrum should be independent on the dimension of the excited region and the output should only increase linearly with the length of the excited region.

The light propagation in the multi-scattering media can be roughly classified into three cases: (i) mean free path $\ell^* \gg \lambda/2\pi$ and λ is the excitation wavelength. It can be thought as an incoherent energy transport [11]. This random walk problem is accurately described by a diffusion equation for the light energy density at large length scale, i.e., $L \gg \ell^*$. (ii) $\ell^* \sim \lambda/2\pi$, the interference of the scattered light becomes very important. Of importance in the strong scattering media are recurrent scattering events, where the light continuously traverses a loop-like a path involving the same set of scatterers. (iii) $\ell^* = \lambda/2\pi$, this is known as the Ioffe–Regel criterion in the small mean free path. Light simply stops flowing over long length scales

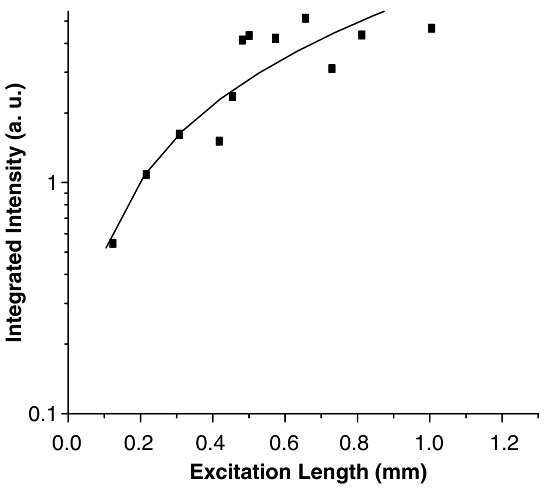

Fig. 6. The dependence of the emission intensity on the excitation stripe length. The solid line is the best fit to the experimental data using Eq. (1).

and Anderson localization of light occurs. In this experiment, the mean free path is $\ell^* = 5.4 \times 10^5$ nm $\approx 1000\lambda$, $k\ell^* \gg 1$, far from Ioffe–Regel criterion. Our case ($k\ell^* \gg 1$) can be described by the Letokhov's theory of light diffusion with gain [12]. If we used the level width $\delta\nu = (c/\lambda^2)\delta\lambda = 7.1 \times 10^{12}$ Hz and level spacing $d\nu/dN = c\lambda^2/8\pi n^3 V = 4.44 \times 10^6$ (Hz), then the Thouless number [13] should be $\delta \equiv \delta\nu/(d\nu/dN) = 1.6 \times 10^6 \gg 1$. It is indicates again that there is no coherent amplification with coherent feedback due to Anderson localization of light in our case.

In summery, the spectral narrowing was demonstrated in (MEH-PPV + TiO_2)/glass waveguide doped with TiO_2 scatterers in a diameter of 20 nm. The lasing emission has obvious direction. As the length of excitation stripe increased, the emission intensity was exponentially enhanced and FWHMs were decreased. The result suggests that ASE is the dominant mechanism for random lasing. The random lasing is far from either Ioffe–Regel criterion of $k\ell^* = 1$ for a passive medium or the Thouless criterion of $\delta = 1$ for an active medium. This suggests that there may be a new criterion suitable to our system.

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