Lasering in a Waveguide with Scatterers in Diameter 20 nm *

LIU Chun-Xu(刘春旭)[1**], LIU Jun-Ye(刘俊业)[1], ZHANG Jia-Hua(张家骅)[1], DOU Kai(窦恺)[2]

[1]*Key Laboratory of Excited State Processes, Changchun Institute of Optics, Fine Mechanics and Physics, Chinese Academy of Sciences, Changchun 130033*

[2]*Department of Physics, Oklahoma State University, Stillwater, OK 74078, USA*

(Received 24 June 2004)

We report random lasing achieved in a MEH-PPV/glass waveguide with the TiO_2 scatterers in diameter 20 nm that is significantly smaller than submicrometre of TiO_2 scatterers in the films or suspensions previously reported on random lasing. The spectral lines are dramatically narrowed by almost two orders of magnitude compared with those excited by a xenon lamp. The amplified spontaneous emission is identified as the dominant mechanism in our system. Light localization might be achieved in a broad class of random materials based on the features of the mean free path $l^* = 5.4 \times 10^5$ nm, $kl^* \gg 1$ and the Thouless number 6.73×10^{-5} with k being the wave number.

PACS: 42. 55. Zz, 72. 15. Rn, 78. 55. Qr

In recent years, the significant advantages of random lasers such as low loss, low cost, high gain, and even a 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 emphasized that laser action is possible in these media due to the process of diffusive feedback[3] and lasing does not rely on the necessary feedback provided by resonant cavities. This type of laser action has been known as the random laser.[4]

The idea for the random laser originally proposed by Letokhov was demonstrated by the experimental discovery of 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 Rhodamine dye when the pump light intensity exceeded the threshold. Pioneering work of random lasing in TiO_2 nanoparticles doped in MEH-PPV/polystyrene films was carried out by Hide *et al.* in 1996.[6]

To our knowledge, the diameters of scatterers at submicrometre have been so far reported for experimental investigation on random lasing. In this Letter, we report, for the first time to our knowledge, the experimental observation of random lasing using the 20-nm TiO_2 scatterers in a 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 by using dip-coating and they contained MEH-PPV1mg/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. Figure 1 shows the emission spectra excited by a xenon lamp with an F-4500 fluorescence spectrometer. The full widths at half maximum (FWHM) of the spectra for films (1 : 0) and (1 : 1) are 124 nm and 103 nm, respectively.

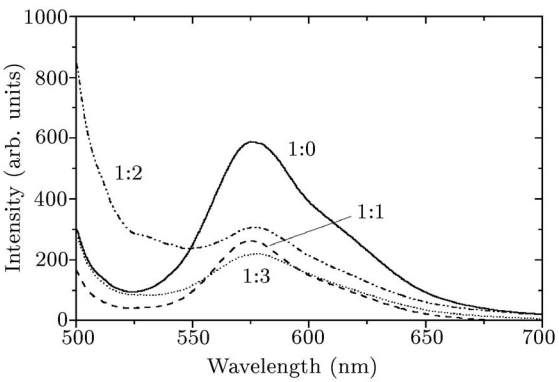


Fig. 1. Emission spectra of the samples excited by a xenon lamp.

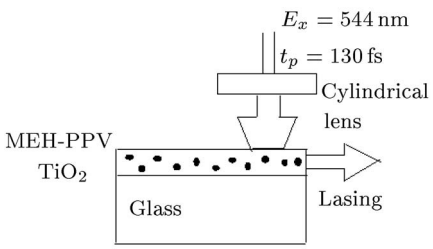


Fig. 2. Schematic of the experimental configuration.

A Spectra-Physics Ti:sapphire laser delivering 3 μJ 150 fs pulses at 544 nm with a repetition rate of 1 kHz

* Supports by the National Natural Science Foundation of China under Grant Nos 10174078, 50172047 and 60308008.

** Email: cxliu@ciomp.ac.cn

©2004 Chinese Physical Society and IOP Publishing Ltd

was used as a excitation source. A schematic diagram for the experimental configuration is shown in Fiq. 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. The lasing action from the current stripe design (one-dimensional gain region) was more easily analysed than the lasing action from a two-dimensional disc. The waveguide structure was found to show only one transverse electric (TE) mode without any transverse magnetic (TM) mode. Most excitations were stimulated to emit into the waveguide modes, and thus a larger fraction of the light came out of the edge.

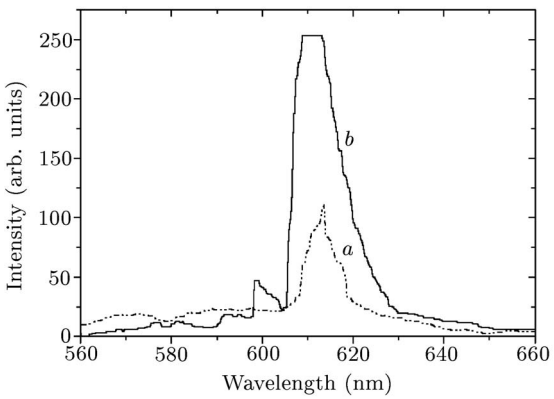


Fig. 3. Emission spectra: (*a*) MEH-PPV (1:0) film; (*b*) MEH-PPV/TiO_2 (1:1) film.

Morphology-dependent resonance (MDR)[7] takes place while the conditions of $d \geq \lambda$ and $n_{\rm sca} > n_{\rm sur}$ are satisfied, where d and λ denote the diameter of the random scatterers and the excitation wavelength, and $n_{\rm sca}$ and $n_{\rm sur}$ represent the refractive index of the scatterers and the surrounding medium. Multi-scattering, possibly associated with the refractive index and size of the scatterers, the 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 submicrometre scatterers in some solutions. For this reason, the 20-nm TiO_2 scatterers were used for the preparation of high quality waveguide adopted in the current experiment. The authors of Ref. [7] came to a conclusion that the scatterers are too small to maintain resonance with high Q-factor. Our results however demonstrate that the lasing actions in the MEH-PPV/TiO_2 (1:1) doped film are much stronger than that in the neat MEH-PPV (1:0) film (Fig. 3) under the same experimental condition as Ref. [7]. The FWHMs of the samples (1:0) and (1:1) are 9 nm and 7 nm, respectively. Amplified spontaneous emission (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 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]

Figure 4 shows the excitation dependence of FWHMs and the integrated intensities emitted from the edge. As excitation intensity is elevated, the FWHMs of spectral lines decrease, whereas the spectral integrated intensity of the edge emitting increases. After the pumping attains 45 μJ/cm^2, the radiation intensity reaches saturation dramatically, which may be due to (i) MEH-PPV:TiO_2/glass configuration and (ii) the measuring system, or other.

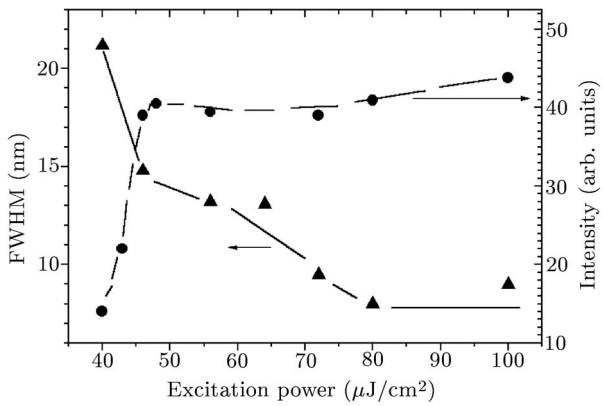


Fig. 4. Dependences of FWHMs of the spectra and the spectrum integrated intensities emitted from the edge on the exciting power of a pulse.

The spectral narrowing mechanisms are still in debate: a few mechanisms of spectral narrowing have been proposed to be cooperative superfluorenscence,[9] bi-excitonic emission,[10] and collective ASE.[8] 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 analysed as follows. Firstly, the spectral lines were broadened at a short length of stripe and the spectral width was found to be narrowed with the increasing length of the excitation stripe (Fig. 5). For various lengths of the excitation stripe, i.e. $\ell = 1.06$, 0.85, 0.76, 0.68, 0.59, 0.51, 0.49, 0.46 and 0.42 mm, we measure the emission spectra to determine the FWHMs that are from 7 to 21 nm for sample (1:1). The FWHM of 21 nm was found to be still narrower than the FWHM 103 nm for $\ell = 0.42$ mm with the same sample using a xenon lamp excitation, as shown in Fig. 2. This implies that the excitation exceeds the intensity threshold. A double-peak emission was also found to appear in the spectra at intermediate pumping levels. Fig. 5 indicates that the lasing modes change with the pumping stripe length.

Secondly, we consider ASE in the case that the

output intensity from one end of the stripe should be given by[8]

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

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 pumping stripe. Figure 6 illustrates the dependence of the emission intensity on the excitation stripe length. The solid line depicts the fit to the experimental data by Eq. (1). If the superfluorescence or biexcitonic emission is of the mechanism responsible for spectral narrowing, the emission spectrum should be independent of the dimension of the excited region and the output should not merely increase linearly with the length of the excited region.

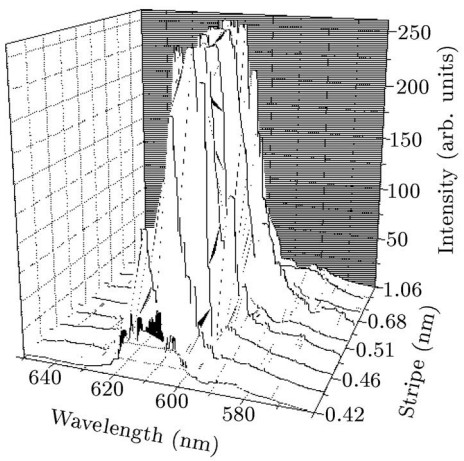


Fig. 5. Emission spectra of the MEH-PPV/TiO_2 (1:1) film under the excitations with various stripe lengths.

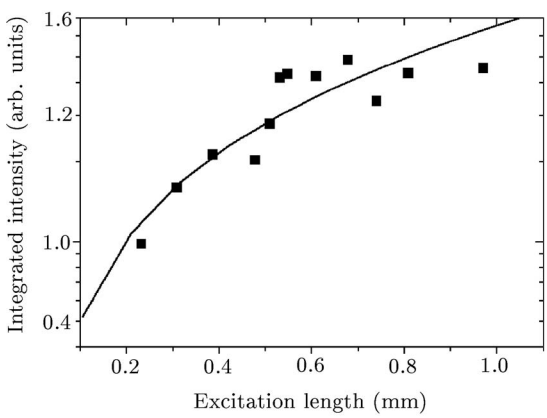


Fig. 6. Dependence of the emission intensity on the excitation stripe length. The solid line is the best fit to the experimental data by Eq. (1).

The light propagation in the multi-scattering media can be roughly classified into three cases: (i) The mean free path $l^* \gg \lambda/2\pi$ with λ being the excitation wavelength. This case can be thought of 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. $L \gg l^*$. (ii) The mean free path $l^* \sim \lambda/2\pi$; the interference of the scattered light becomes very important. Of importance in the strong scattering media is the recurrent scattering event, where the light continuously traverses a loop-like path involving the same set of scatterers. (iii) $l^* = \lambda/2\pi$; this is known as the Ioffe–Regel criterion in the small mean free path. Light simply stops flowing over long length scales and the Anderson localization of light occurs. In this experiment, the mean free path is $l* = 5.4 \times 10^5$ nm $\alpha 1000\lambda$, $kl^* \gg 1$, far from the Ioffe–Regel criterion. This indicates that the random lasing has little to do with the Anderson localization of light in our experiment.[12] However, recently it is reported that even though $k\ell^* = 32 \gg 1$, coherent feedback due to the Anderson localization of light can still occur.[13] It is well known that the coherent back scattering or the Anderson localization and coherent amplification in a disorder medium are two bases for the random laser. On the other hand, the Thouless number can be calculated by $\delta \equiv \delta\nu/(d\nu/dN) = 6.73 \times 10^{-5} \ll 1$, where $\delta\nu$ is the width of energy level, $d\nu/dN$ is the level spacing.[14] The Thouless number $\delta = 1$ is thus unsuitable to our experiment.

In summary, the spectral narrowing has been demonstrated in a (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 increases, the emission intensity is exponentially enhanced and the FWHMs decrease. The result suggests that ASE is the dominant mechanism for random lasing. The random lasing is far from either the Ioffe–Regel criterion of $kl^* = 1$ 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.

References

[1] Daozhong Zhang, Bingying Cheng et al 1995 *Opt. Commun.* **118** 462
[2] Siddique M and Alfano R 1996 *Opt. Lett.* **21** 450
[3] Letokhov V S 1968 *Sov. Phys. JETP* **26** 835
[4] Wiersma D S, van Albada M P et al 1995 *Nature* **373** 203
[5] Lawandy N M, Balachandran R M et al 1994 *Nature* **368** 436
[6] Hide F B, Schwartz J M et al 1996 *Chem. Phys. Lett.* **256** 424
[7] Oliveira P and Lawandy N 1997 *Opt. Lett.* **22** 895
[8] McGehee M, Gupta R et al 1998 *Phys. Rev.* B **58** 7035
[9] Pradhan P and Kumar N 1994 *Phys. Rev.* B **50** 9644
[10] Jeoung S C et al 1999 *Appl., Phys. Lett.* **74** 212
[11] Prasad B R et al 1997 *Appl. Opt.* **36** 7718
[12] Gelinik G et al 1997 *Chem. Phys. Lett.* **265** 320
[13] Cao H et al 2002 *Phys. Rev.* E **66** R025601
[14] Cao H et al 2000 *Phys. Rev. Lett.* **24** 5584