Random Fiber Laser

Christiano J. S. de Matos,[1,*] Leonardo de S. Menezes,[2] Antônio M. Brito-Silva,[3] M. A. Martinez Gámez,[4] Anderson S. L. Gomes,[2] and Cid B. de Araújo[2]

[1]Programa de Pós-Graduação em Engenharia Elétrica and Faculdade de Computação e Informática, Universidade Presbiteriana Mackenzie, São Paulo-SP, 01302-907, Brazil
[2]Departamento de Física, Universidade Federal de Pernambuco, Recife-PE, 50670-901, Brazil
[3]Programa de Pós-Graduação em Ciência de Materiais, Universidade Federal de Pernambuco, Recife-PE, 50670-901 Brazil
[4]Centro de Investigaciones en Optica, C.P. 37150 Leon, Gto., Mexico

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We investigate the effects of two-dimensional confinement on the lasing properties of a classical random laser system operating in the incoherent feedback (diffusive) regime. A suspension of 250 nm rutile (TiO_2) particles in a rhodamine 6G solution was inserted into the hollow core of a photonic crystal fiber generating the first random fiber laser and a novel quasi-one-dimensional random laser geometry. A comparison with similar systems in bulk format shows that the random fiber laser presents an efficiency that is at least 2 orders of magnitude higher.

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The emission of laserlike radiation in highly scattering gain media [1,2], known as random lasers (RLs), has received considerable attention for over a decade due to its unique properties and to a number of potential applications [2]. The optical feedback in RLs is provided within the amplifying medium by random light paths that result in an increased emission intensity and reduced emission linewidth. The RL action can take place in two distinct regimes [2]. When the length of the scattering gain medium is greater than the photon mean free path, and this path, in turn, is greater than the emission wavelength, light propagation is diffusive. In this case, the probabilistic nature of diffusion means that interference contributes negligibly to the feedback process which is incoherent. On the other hand, if the photon mean free path and the emission wavelength have the same order of magnitude, localization of the radiation field occurs within the structure and coherent feedback is possible [3].

An important concept for RLs is that of the β factor, defined (both for conventional and random lasers) as the fraction of spontaneous emission that seeds the laser process [4]. The value of β is directly related to the sharpness of the laser threshold, with a thresholdless laser having $\beta = 1$. While in conventional lasers β depends on both the fraction of emitted photons that is collected by the cavity mirrors and the fraction of the spontaneous emission spectrum that overlaps with the laser spectrum, in RLs β depends only on the latter. Thus, β in RLs is usually considerably larger. However, the lack of directionality in these devices has largely limited their application as high β sources.

Recently, a system consisting of a stack of reflecting glass slides of variable thicknesses alternated with amplifying layers was presented [5]. This system was shown to behave as a one-dimensional random laser in the strong localization regime, in which pump and emission longitudinal modes arise deep in the sample. When these two modes overlap with each other and with the amplifying layers, a very low RL threshold is obtained and the RL action is very efficient. Such an approach, however, is not applicable to the diffusive (incoherent feedback) RL regime, in which stable longitudinal modes are never formed. It is worth noting that a theoretical model for 1D RLs operating in the strong localization regime was also reported [6].

A configuration that is extensively used for random lasing with incoherent feedback was demonstrated by Lawandy *et al.* [1], in which subwavelength scatterers are suspended in a dye solution. Pump scattering restricts population inversion to a thin disk close to the solution surface [7]. This means that photons can diffuse out of the active region, contributing to loss and to an increase in lasing threshold. Decreasing this threshold generally requires an increase in dye and/or scatterer concentration. Alternative methods are able to increase the lasing efficiency, e.g., providing external feedback with a mirror that increases the photon lifetime in the solution and, thus, decreases the threshold pump energy [8] or using surface plasmon excitation to locally increase the radiation field and to reduce the lasing threshold [9].

Both directionality and increased efficiency can be expected if the scattering gain medium is inside a waveguide that transversely confines light. A 1-mm-thick planar waveguide RL where liquid crystal droplets acted as scatterers was recently demonstrated [10] and a reduction in the pump intensity threshold and a decrease in the emission linewidth by 33% were observed. It is clear that a channel waveguide, providing two-dimensional confinement, would further improve the RL characteristics. However, such a configuration has not yet been investigated for RLs in any regime, possibly due to the fact that a practical cladding material, presenting a refractive index that is lower than that of the scattering gain solution, was unavailable.

The advent of hollow-core photonic crystal fibers (PCFs) [11] opens new possibilities. In PCFs the microstructured cladding presents an effective refractive index that is a weighted average between the indices of glass and air and may have values below 1.3. In addition, hollow-core PCFs may present photonic-band gap guidance, which confines light to the core irrespective of its refractive index. These features have enabled the design, fabrication, and application of liquid-core PCFs [12,13]. Of particular importance here is the demonstration of fiber dye lasers [13], obtained when PCFs were filled with a dye solution and end-pumped to yield conventional laser action.

In this Letter, we demonstrate random laser action in a channel waveguide geometry consisting of a hollow-core PCF. The PCF (Crystal Fibre A/S, model HC-1550-02) had core diameter, cladding pitch and cladding air-filling fraction of $\sim$10.9 μm, 3.8 μm and >90%, respectively, and its cross-sectional profile is depicted in the inset of Fig. 1. The scattering gain medium, consisting of a suspension of 250-nm rutile (TiO_2) in a solution of Rhodamine (Rh) 6G in ethylene glycol, was selectively inserted into the core, leaving the cladding microstructure filled with air. The dye and scatterer concentrations were varied in the range 10^{-5} mol/l $\leq \rho_{\rm dye} \leq 10^{-3}$ mol/l and 10^{7} cm$^{-3} \leq \rho_{\rm scatt} \leq 10^{9}$ cm^{-3}, respectively. For comparison, Rh 6G solutions without rutile particles were also investigated.

As the cladding effective refractive index was numerically estimated to be 1.27 and the refractive index of ethylene glycol is $\sim$1.43, the PCF core was able to confine and guide light through total internal reflection. For $\rho_{\rm scatt} = 10^{8}$ cm^{-3}, the average distance between scatterers is 21 μm. As the mean free path of photons within the suspension is expected to be a few times larger than this average distance, scattering events are unlikely to occur for photons that radially cross the PCF core. Consequently, total internal reflection is the main mechanism transversely confining light within the gain medium. Scattering events mainly occur along the fiber axis, which results, to the best of our knowledge, in the first quasi-one-dimensional (1D) RL reported in the diffusive regime. Also, we note that transverse confinement is not obtained in the 1D RL presented in [5].

To insert the liquid gain medium into the core of a $\sim$50-mm-long piece of PCF while leaving the cladding holes filled with air, we followed the procedure described in [14] and used a 3 ml disposable syringe to press the liquid into the fiber core. The PCF was subsequently removed from the syringe and cleaved.

The experimental setup for the optical measurements is shown in Fig. 1. The second harmonic of a Q-switched Nd:YAG laser (532 nm, 7 ns, 5 Hz) was used to laterally pump the PCF. An iris with a 4 mm diameter aperture is used for obtaining a top-hat beam which is focused by a 50 mm focal length cylindrical lens on the final portion of the PCF, so that a 50 μm $\times$ 4 mm area of the PCF receives nearly uniform illumination. Because of the small core diameter, all the dye molecules within the 4-mm section are equally excited, and not a thin superficial layer as in previous schemes. The maximum intensity (fluence) inside the PCF core was 144 MW/cm^2 (2.1 J/cm^2). The light emitted along the PCF axis was then collected by a 0.44 N.A. optics and sent to a spectrometer. Care was taken to align the cylindrical lens with the PCF's axis, since it was observed that the recorded spectra substantially vary with this alignment. We also controlled the laser intensity to prevent bleaching of the dye molecules.

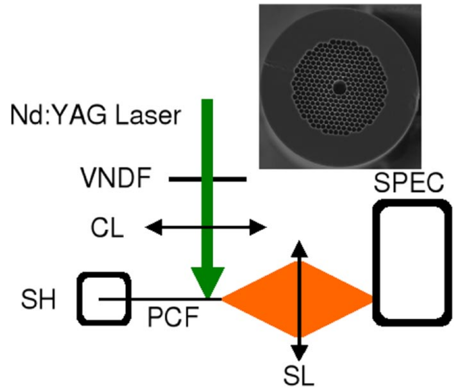

FIG. 1 (color online). Side view of the experimental setup. Variable neutral density filter (VNDF); 50 mm focal length cylindrical lens (CL); Sample holder (SH); 50 mm focal length spherical lens (SL); Spectrometer (SPEC). Inset: Scanning electron microscopy of the photonic crystal fiber used in the experiment (provided by Crystal Fibre A/S).

Figure 2 displays the emission spectra for various pump intensities and for different configurations. Figure 2(a) shows results for the conventional RL scheme for a suspension with $\rho_{\rm dye} = 10^{-4}$ M and $\rho_{\rm scatt} = 10^{8}$ cm^{-3} placed inside a cylindrical quartz cuvette (radius = height = 10 mm). For a fair comparison with results obtained with the random fiber laser setup, pumping in the present case also occurred along a 4-mm line. The emission linewidth remains almost constant when the pump intensity is increased from 9 to 144 MW/cm^2. The not so broad linewidths measured ($\sim$22 nm FWHM), as compared to the results reported in the literature [1,9,15], occur because the optics at the fiber output is optimized to collect light from the pumped region and not from the unpumped surrounding, where photon absorption and reemission broadens the linewidth. In Fig. 2(b), the spectra are shown for a medium with $\rho_{\rm dye} = 10^{-4}$ M and $\rho_{\rm scatt} = 0$ cm^{-3}, placed inside the PCF core. Again, it is observed that the emission spectral linewidth does not change much for the same pump intensity range, but in this case the spectra are narrower than in the previous situation. This is due to the reduced transverse dimensions inside the PCF, as compared to the bulk case, that further prevents the absorption and emission of Stokes photons with smaller energies that contribute for broadening of the emission spectrum.

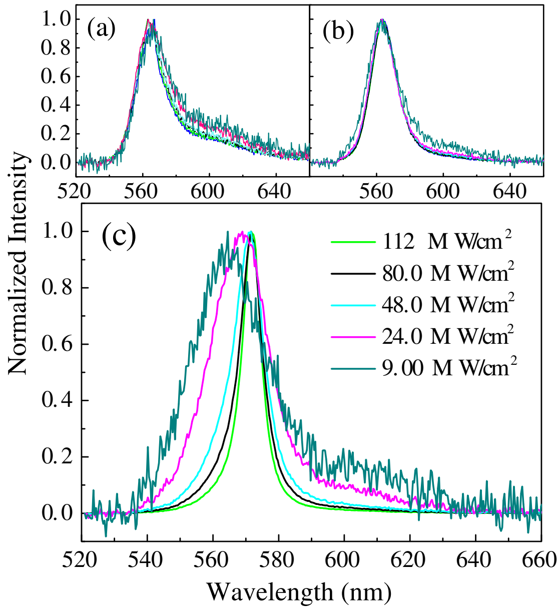

FIG. 2 (color online). Emission spectra measured along the excitation region for different pump intensities and different configurations. (a) A 10^{-4} M solution of Rh 6G in ethylene glycol containing 10^8 cm^{-3} rutile particles inside a cell. (b) A 10^{-4} M solution of Rh 6G in ethylene glycol inside the photonic crystal fiber, without scattering particles. (c) A 10^{-4} M solution of Rh 6G in ethylene glycol inside the photonic crystal fiber with 10^8 cm^{-3} rutile particles.

Finally, Fig. 2(c) shows the results with rutile particles inside the PCF. The solution into the core had $\rho_{dye} = 10^{-4}$ M and $\rho_{scatt} = 10^8$ cm^{-3}. The linewidth significantly decreases when the pump intensity increases, demonstrating that the presence of scatterers is essential for observing spectral line narrowing. The emission redshift as the pump intensity is increased has been previously observed [16] and explained [17]. In the present case, for low pump intensities, spontaneous emission dominates and detected photons are mostly emitted from the region closest to the fiber output. As pump intensity is increased the higher gain allows for photons emitted further into the fiber to reach the output. The long path within the gain medium then increases photon absorption and reemission, causing the redshift [17].

Figure 3 shows the emission peak intensity and spectral linewidth (FWHM) as functions of the pump intensity for the cases shown in Figs. 2(b) and 2(c). When rutile is present, a large increase in the emission peak intensity is observed for intensities larger than the threshold pump intensity ($I_{thr} \sim 40$ MW/cm^2). Unlike bulk RLs, the random fiber laser presented a threshold behavior even if the emission intensity was integrated over its spectrum, as shown by the triangles in Fig. 3(a). As pointed in Refs. [4,6], such a behavior indicates that the threshold is not only a result of spectral narrowing, but also of the existence of directionality in the emission above threshold. The observed threshold behavior is, therefore, evidence

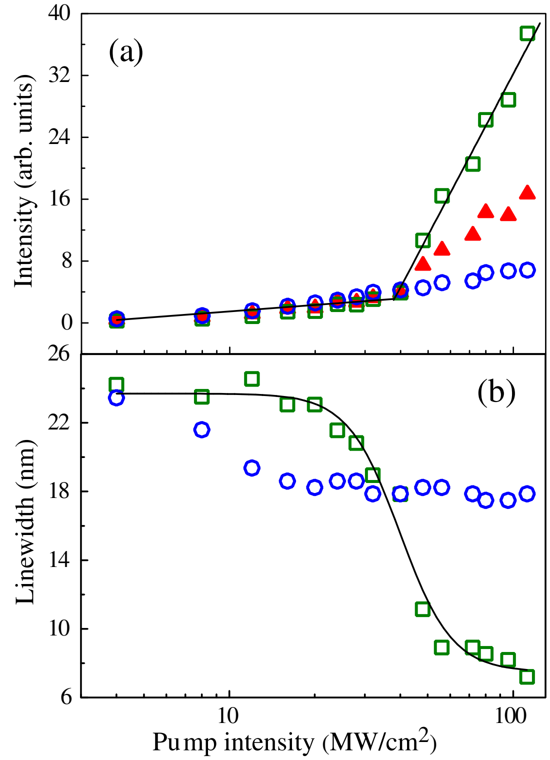

FIG. 3 (color online). (a) Emission peak (open symbols) and spectrum-integrated (solid symbols) intensities for 10^{-4} M Rh 6G in ethylene glycol with (squares/triangles) and without (circles) 10^8 cm^{-3} rutile particles in the core of the PCF. (b) Emission linewidth (FWHM) for the same configurations. Symbols represent the same conditions as in (a). The solid lines are guides to the eye.

that the emission in the random fiber laser is directional. It can also be seen that when rutile was not present in the PCF core (circles) no intensity threshold behavior could be detected. Figure 3(b) shows that at threshold the emission linewidth reduces from ~24 nm to ~7 nm, which is typical of RL action. The linewidth dependence on pump intensity when a Rh solution (without scatters) is inside the PCF is also depicted. Note that no laser threshold is observed. The slight reduction in linewidth, which has also been observed in [1], is believed to be the effect of the spectral dependence of gain, which for high gain values naturally causes a narrowing of the amplified spontaneous emission, especially in long gain media. The evidence described above strongly indicates that random lasing was achieved for the first time within a PCF. The comparison with the bulk sample corroborates this conclusion and practically rules out other explanations based on, e.g., saturable absorption.

To compare our results with other reported RLs, we define a figure of merit $\mathrm{FOM} = (I_{thr} \cdot \rho_{dye} \cdot \rho_{scatt})^{-1}$. In the seminal paper by Lawandy *et al.* [1] $\mathrm{FOM} = 1.3 \times 10^{-8}$ cm^5 1/MW · mol. In Ref. [18], the authors investigate the temporal and spectral behavior of a RL for different values of ρ_{dye} and ρ_{scatt}, obtaining at best

FOM $= 1.1 \times 10^{-9}$ cm^5 l/MW $\cdot$ mol. RL systems in which both the dye molecules and the scattering particles were imbedded in a polymeric host were also studied [15] and the results lead to FOM $= 1.7 \times 10^{-9}$ cm^5 l/MW $\cdot$ mol. The use of external feedback to decrease the pump intensity threshold [8] was investigated in a suspension of TiO_2 in a Rh 640 solution, with results leading to FOM $\sim 3.5 \times 10^{-8}$ cm^5 l/MW $\cdot$ mol. In Ref. [19], the authors investigated RL action of a dye solution containing aggregates of $\sim$700 nm in diameter formed by $\sim$100 nm in length TiO_2 rods. In this case, the concentration of scattering particles is not given, but assuming the same concentration as in the present Letter, one gets FOM $= 4.4 \times 10^{-8}$ cm^5 l/MW $\cdot$ mol. More recently [20], another RL scheme, using 55 nm in diameter silver nanoparticles instead of TiO_2 particles as scattering centers, was introduced having FOM $= 6.1 \times 10^{-12}$ cm^5 l/MW $\cdot$ mol. In the present work, we obtain FOM $= 2.5 \times 10^{-6}$ cm^5 l/MW $\cdot$ mol. The significantly higher efficiency obtained here in comparison with the previously reported results is due to the novel channel waveguide RL geometry, that favors a transverse feedback mechanism contributing to an increase of the photon lifetime within the scattering gain medium. A comparison with the 1D RL studied in [5] is not straightforward due to the substantial differences in the construction of that system. Nevertheless, assuming ρ_{dye} in that case to be the inverse of the cube of the average distance between reflecting surfaces, one obtains FOM $= 2.9 \times 10^{-3}$ cm^5 l/MW $\cdot$ mol. This value is substantially higher than that of the present work, but is achieved due to the emergence of longitudinal modes in the strong localization regime. In contrast, the channel waveguide RL approach is applicable to RLs in any regime and is of particular importance for diffusive RLs.

The transverse feedback results in peculiar characteristics that are not present in other RL configurations. First, directionality, which is an attractive feature for some potential applications, is naturally obtained. Second, although a specific characterization was not performed, the multiple lateral reflections are expected to generate transverse laser modes. This feature is a direct consequence of the studied configuration not being a RL in the transverse direction. Using the ethylene glycol refractive index and the effective cladding index, the liquid-core PCF numerical aperture is calculated to be $\sim$0.657, which from simple waveguide estimates [21] results in over 700 guided modes. In a ray optics approximation, the ray corresponding to the highest-order guided mode makes a 27° angle with the fiber axis. Note, nevertheless, that the scattering events are expected to result in a random coupling between the guided modes.

To derive an estimate for β in the random fiber laser, we used the method presented in [16], multiplying the result by the fraction of the isotropically emitted photons that are collected by the fiber core, to account for directionality. We obtain $\beta = 1.4 \times 10^{-2}$, which is 10 times smaller than typical values for bulk RLs but is still significantly higher than those for most conventional lasers [4,16]. The demonstrated device, therefore, presents a compromise between a high beta value and directional emission, occupying a so far unexplored region in the laser-parameter space.

In conclusion, we reported the operation of a quasi 1D RL in the diffusive regime, using a PCF as a confining waveguide. Because of the light confinement properties of this structure, the RL action is more efficient than that obtained in similar RLs in bulk format previously reported in the literature. For the full understanding of this system, a theoretical model, which also considers the possibility of obtaining single-mode random lasing in a fiber geometry, will be presented in the near future.

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*Corresponding author.
cjsdematos@mackenzie.br

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