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Random Lasers: review of research activities at IPEN

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***Abstract*— Random lasers offer advantages such as low-cost fabrication and robustness in harsh environments and have applications in sensing, imaging, communications, and security. Our current research focuses on advanced materials, active control techniques, integration with other photonic structures, and exploration of Anderson localization and polydisperse effects. Coherent feedback in cavity-enhanced random lasers is discussed as means to achieve specific emission characteristics, and emission at 1300 nm is shown for the first time.**

Keywords—random laser, Anderson localization, aerogel, glass

I. Introduction

Random lasers (RLs) are a type of laser based on the principle of multiple scattering of light in a disordered or random medium, rather than relying on conventional resonant cavities. Unlike conventional lasers, random lasers do not require mirrors or other optical components to form a resonant cavity for feedback. In a random laser, the active medium, which can be a solid, liquid or gas, contains scattering centers or disordered structures that scatter light in multiple directions. This scattering process creates a random path for light propagation in the medium. When the active medium is excited by an external energy source, such as optical pumping or an electrical discharge, the light is amplified as it is repeatedly scattered and travels through the medium. Eventually, some of the photons are excited to emit, forming coherent laser light. RLs generally show a lack of well-defined resonator modes, with some notable exceptions, instead, they exhibit broadband emission spectra and are highly diffusible. The output of a random laser can be highly directional or diffuse, depending on the specific design and properties of the active medium. RLs have been investigated for applications in imaging, sensing, communications, and lighting.

Random lasers offer unique characteristics such as low-cost fabrication, broad emission spectra and robustness in challenging environments, making them suitable for various applications such as sensing and detection, where they are used in chemical and biological sensing, environmental monitoring of pollutants and hazardous substances and structural health monitoring of buildings and bridges. Other applications are in imaging, where Random lasers are used in fluorescence microscopy, speckle-free imaging systems, and metrology for distance measurement and interferometry. Another application is in security and authentication, where random lasers can be used in anti-counterfeiting measures and authentication systems, providing secure and tamper-proof labeling and tagging. The latter applications are due to the unique spectral and spatial distribution, and spatially complex emission that provides a distinct and unique signature making them inherently difficult to replicate or mimic.

Random lasers are an open research frontier and are being explored to enhance their performance, develop new functionalities, and expand their applications. Some of the emerging frontiers for random lasers include advanced materials through tailored gain materials for improved performance and extremely low threshold; active control through developing techniques for active manipulation and optimization of gain and spectral characteristics; hybrid systems by integrating random lasers with other photonic structures such as on-chip photonic platforms; nonlinear effects for enhanced functionality and quantum Random Lasers based on quantum gain media such as quantum dots and quantum cascade random lasers.

II. Random lasers studies

A. *Directional Random Lasers with coherent feedback*

Directional random lasers with coherent feedback have potential applications in various fields, including imaging,

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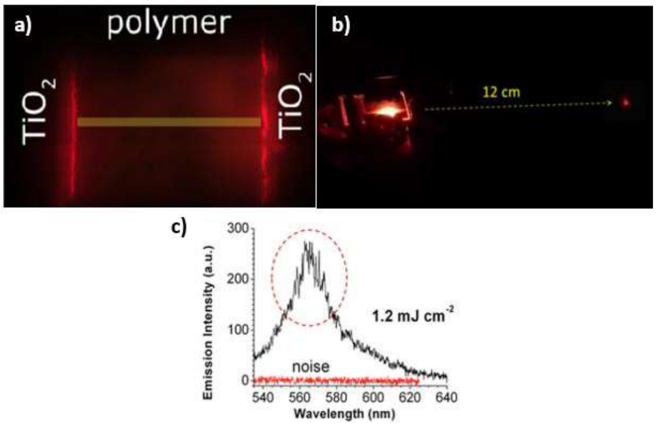

Figure 1: a) Image of the polymer strip with TiO2 particles. b) Low divergence red laser emission of microfluidic RL. c) Persistent, localized modes on top of the ASE band of the extended modes, indicating suppression of the interaction between the localized modes.

sensing, communication, distance measurement and interferometry [1, 2]. Their ability to generate controlled laser emission without the need for conventional cavity structures makes them attractive for applications where simplicity, compactness, and low-cost are desired.

In a first embodiment of such a laser in 2015, we enclosed an active region, a DCM doped DNA-CTMA polymer film, between two rough scattering surfaces, where scattering centers were randomly positioned. No distributed feedback inside the active material itself was imposed, the scattering regions being positioned at the edges of the pumped area [3]. The entire device was acting as a single random oscillator with dimensions of several hundreds of micrometers, being a promising device for lighting, display and imaging applications (Figure 1a).

In a next step, we integrated a directional RL into an optofluidic device using a hollow-core antiresonant reflecting optical waveguide (HC-ARROW) containing the gain media inside a reservoir, which was connected to microchannel waveguides to increase beam directionality [4]. With the reservoir and microchannels, a low laser threshold of below 100 µJ and a high radiance with beam divergence of 68 mrad, comparable to a traditional Nd:YAG laser, were achieved, aside from a much slower dye decay rate (Figure 1b).

B. *Anderson localization and Random Lasers*

Combining Anderson localization and random lasers makes much sense: Anderson localization is a phenomenon where wave propagation is strongly affected by disorder, leading to the confinement of waves in a localized region. Random lasers, on the other hand, utilize multiple scattering within a disordered medium to achieve lasing action. The interplay between disorder-induced localization and lasing opens up new avenues for designing and manipulating light sources with specific properties, such as highly localized emission, narrow linewidths, or tunable spatial patterns. It also offers opportunities for studying fundamental physics related to wave localization and lasing in complex media [5, 6]. We investigated the random laser action at the transition to Anderson localization in a strongly disordered scattering medium composed of a colloidal suspension of core−shell nanoparticles (TiO_2@Silica) in ethanol solution of Rhodamine 6G [7]. Narrow peaks with a linewidth of $\leqslant 0.17$ nm and of approximately equal intensity were observed on top of the classical superfluorescence band (ASE, amplified spontaneous emission), indicating suppression of the interaction between the peaks modes (Figure 1c). The linewidth of the coherent localized modes is much smaller than the linewidth of the extended modes, which is approximately 1.3 nm, demonstrating the very high Q factor of the localizing cavities. Random laser threshold is also very small, of the order of 10 µJ mm^{-2} [8].

Correlation-induced localization is an active area of research, and the specific requirements for inducing and controlling localization is determined by a combination of factors related to the system's disorder, scattering properties, and the desired level of localization. For example, if the scatterers are arranged in a highly ordered periodic pattern, the localization effect may be diminished compared to a disordered arrangement [9]. The correlation length represents the characteristic distance over which the scattering events are correlated. When the correlation length is comparable to the wavelength of light, the localization effect becomes more pronounced. By increasing the shell thickness in the core-shell nanoparticles from 40 nm (TiO2@SiO_2) to 70 nm (TiO_2@SiO_2-SiO_2), a stronger and longer-range Coulomb interaction between the scatterers was induced, increasing correlation (Figure 2a) and, as a consequence, localization and slope efficiency (Figure 2b) [10].

C. *Polydisperse Random lasers*

The presence of different-sized scatterers in the medium can enhance scattering and light trapping effects. The multiple scattering events and the variety of scattering cross-sections from different scatterers can increase the path length

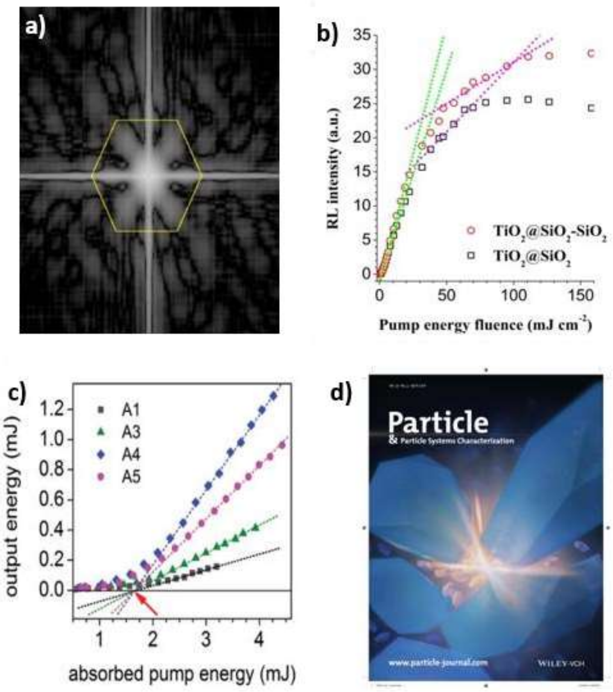

Figure 2: a) diffracted light collected in the backscattering configuration for the RL systems (TiO_2@SiO_2–SiO_2). b) a higher contribution of RL localized modes for the TiO_2@SiO_2–SiO_2 RL system is detected when compared to the TiO_2@SiO_2 RL system, as shown by the higher slope (red circles, green slope), accompanied by a 25% higher pumping fluency for saturation of the RL. c) Output energy as a function of absorbed pump power for polydispersed samples. A1 to A5 have increasingly higher mean diameter, but all have the same threshold (indicated by arrow) demonstrating that the smaller particles are responsible for laser threshold. d) artist impression of the small particles concentrating the light.

and the interaction of light with the gain medium, leading to improved energy deposition and amplification. By preparing pressed pellets of Nd^{3+}:YVO_4 powder, which has a very high gain [11, 12], and optimizing them in terms of grain size distributions, we achieved an increase of 130% (2.3x) in output power, with respect to the best monodisperse powder [13]. A maximum output pulse energy of 1.3 mJ was achieved with a record slope efficiency of 50% as a function of absorbed pump power (figure 2c), demonstrating that these random lasers can be efficiently pumped by semiconductor diodes. Light diffusion in these RLs is governed mainly by the larger particles and it is almost independent of the smaller particles, whereas the smaller particles, which are trapped between the larger particles, are responsible for an increase of 15% in the number of scattering events, increasing the local pump power density by up to five times (an artist impression of this behavior is shown in figure 2d).

D. Cavity-enhanced coherent random lasers

Coherent feedback in random lasers is influenced by the interplay between gain and scattering. The gain medium should provide sufficient amplification to compensate for scattering losses and allow the light to build up coherently within the system. Adjustment of the gain profile and optimization of the scattering strength, anisotropy and spatial distribution of the scatterers are necessary to control multiple scattering events and improve coherence. The spectral emission of a Rhodamine doped aerogel random laser was unequivocally associated with coherent emission, facilitated by an effective cavity of length 67 μm, formed by the combination of the input sample surface and the interface between gain and reabsorption within the pumped sample [14]. This laser system, characterized as weakly scattering ($l_s > l_a$) with an absorption length of 53 μm and a scattering length of approximately 6 mm, demonstrated that the free spectral range (FSR) of the coherent emission peaks (figure 3a), occurring near the laser threshold, corresponded to a cavity length about 25% longer than the absorption length l_a. Above threshold, one single broad emission peak, attributed to amplified spontaneous emission (ASE), was observed.

In the following new embodiment of a cavity-enhanced random laser, we show for the first time a glass random laser, based on neodymium doped zinc tellurite and aluminum oxide (figure 3b) heavy-metal glass [15-18], that operates at 1300 nm (figure 3c) [19]. This laser system works, unlike the weakly scattering system above, in the strongly scattering regime ($l_a > l_s$) with approximately the same absorption length as above (65 μm) and a much smaller transport mean free path of just 4 μm. Being a glass laser of inhomogeneous spectral broadening, no peaks were detected near threshold, but two different slopes were observed (figure 3d), the first attributed to emission from cavity modes, whereas the second is the combination of ASE and coherent emission. A cavity consisting of the pump surface of the sample and an effective mirror formed by a second parallel layer at the gain-loss interface (a combination of absorption and scattering) is probably the main lasing mechanism of this random laser system, similar to the weakly scattering system described above, with the difference that the limiting parameter for the cavity length is the short transport mean free path, in contrast to above where the limiting factor is the short absorption length.

The reason for the lack of emission at 1064 nm is believed to be a measured temperature rise in the active

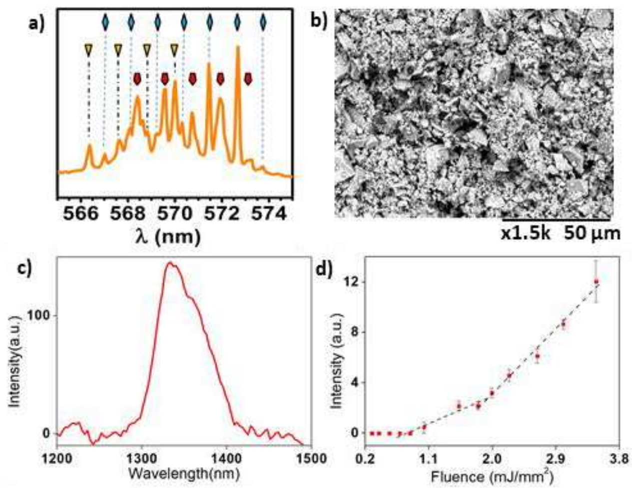

Figure 3: a) three sets of equidistant peaks with the same FSR corresponding to three resonators of the same length within the pump spot size. b) image of the random laser surface. c) emission at 1337 nm. d) output power versus input power clearly showing two different slopes, the first attributed to cavity-enhanced coherent emission and the second to additional incoherent emission due to ASE or extended modes.

volume of the sample, whose loosely arranged and tiny micrometer-sized particles heat up sufficiently during the short pump pulse duration to cause significant population occupation of the lower laser level, hampering inversion at the 1064 nm transition.

Both examples of random lasers, in the weak and strong scattering regimes, demonstrate a wealth of new spectral features, such as controllable equidistant peaks and new wavelengths, that underline their usefulness in the aforementioned, diverse applications.

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