Universal Properties of Random Lasers

Randall C. Polson, Mikhail E. Raikh, and Z. Valy Vardeny

Abstract—The design and fabrication of laser resonators is often difficult. However, random lasers occur in gain media with numerous scatterers and produce coherent laser emission without any predesigned cavity. The generation of coherent emission from multiple scattering is quite general and its basic principles are shown here using two model systems, namely π-conjugated polymer films and rhodamine-TiO_2 suspensions. Above a threshold excitation intensity, both systems show narrow emission lines (<0.5 nm), coherence that is determined by photon statistics, and a fundamental cavity length in the disordered material that is revealed by averaging multiple power Fourier transform spectra.

Index Terms—Fourier transform, laser resonator, random media.

I. INTRODUCTION

RANDOM LASING is a blanket term to cover systems where laser emission is produced from a random collection of scatterers in a gain medium. A diverse number of systems has shown this behavior including neodymium-glass powders [1], dye-TiO_2 solutions [2], nanoclusters of ZnO [3], π-conjugated polymer films [4], dye infiltrated opals [5], dye-TiO_2 polymer films [6], and laser dye within liquid crystals [7]. Properties normally associated with laser emission are all present in these random lasers, including coherence and narrow emission lines. Here, we discuss the properties of random lasers by summarizing recent work with two model systems, namely polymer films and suspensions of scatterers with optical gain.

II. LASER EMISSION PROPERTIES

There is a typical progression of the emission characteristic properties from gain media, from a broad photoluminenscenese spectrum at low excitation intensities, to a narrow stimulated emission band at higher excitation intensities, to narrow emission lines at even higher excitation intensities. Fig. 1 shows these three stages for a dye-TiO_2 suspension. The inset of Fig. 1 shows the typical photoluminescence spectrum, which upon higher excitation collapses into a narrow amplified spontaneous emission spectrum. At even higher excitation very narrow emission peaks (<0.5 nm) appear in the emission spectrum. The characterization and origin of these narrow lines is the goal of this paper.

There are several experimental details that are used to observe the very narrow emission lines from a TiO_2-rhodamine dye suspension. The dye is at a concentration of 3×10^{-2} mole/liter, whereas the 300-nm diameter spherical TiO_2 particles are at a concentration of 8×10^9 particles/cm^3. The dye suspension
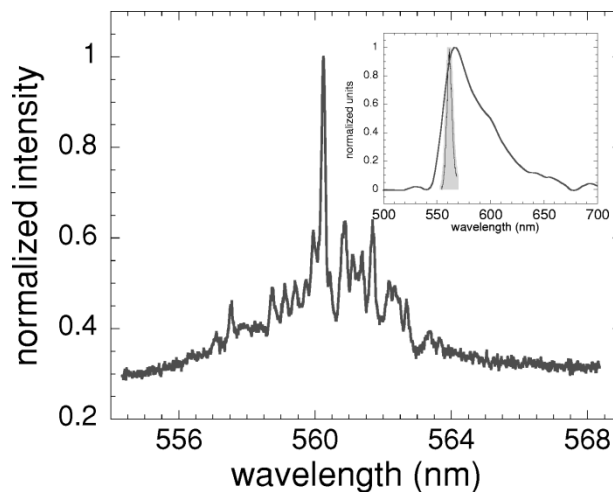
Fig. 1. Rhodamine 6G and TiO_2 suspension showing narrow emission lines. The inset is the regular photoluminescence and spectral narrowing due to amplified spontaneous emission (ASE), shown on a different scale.
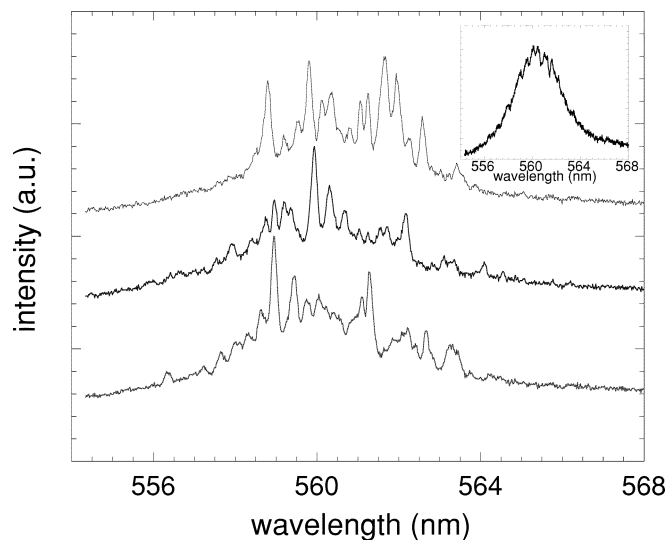

Fig. 2. Three laser emission spectra for a dye and scatterer suspension each excited with a single pulse, shown with an offset for clarity. The inset shows the averaged spectrum of 100 excitation pulses.

is excited with the second harmonic of a Nd:YAG regenerative amplifier at 532 nm, with 100-ps pulses, at a repetition rate of 100 Hz. The excitation is sent through a cylindrical lens and the emission is collected from the long side of the stripe using a fiber optic; it is then sent to a 0.5-m spectrometer, detected with a charge coupled device (CCD) camera and recorded with a PC.

The "spiky" spectrum seen in Fig. 1 was the recording of *one* excitation pulse. Since the scatterers are in motion in the suspension, each excitation pulse illuminates a different configuration of scatterers. Fig. 2 shows the spectra from three different pulses; though the spectra are similar they are *not the same*. If
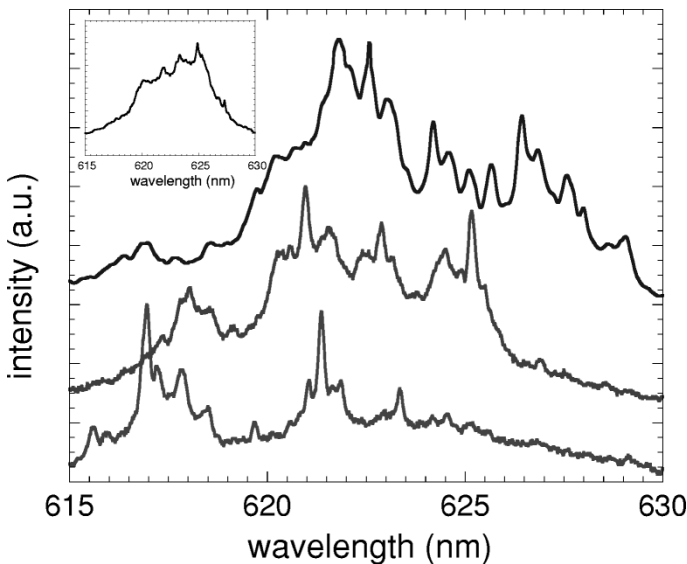
Fig. 3. Three laser spectra for a $DOO - PPV$ polymer film with excitation area of 1 by 100-μm. The inset is a spectrum with a 6 by 100-μm excitation area.
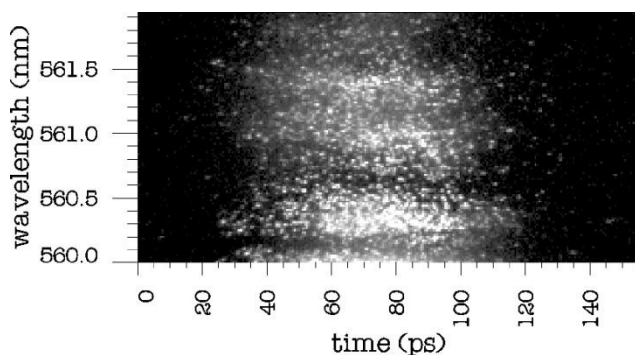

Fig. 4. Time resolved spectroscopy of rhodamine and TiO_2 scatterers suspension obtained with a single excitation pulse.

many excitation pulses are collected, then the resulting spectrum is smooth, as seen in the inset of Fig. 2 where the combined spectrum of 100 separate excitation pulses is shown. We conjecture that the recorded spectrum shows more pronounced peaks with fewer excitation pulses.

Very similar emission spectra characteristics are observed with a π-conjugated polymer film. The specific polymer was poly(dioctyloxy) phenylene vinylene, $DOO - PPV$. The same experimental setup was used as with the dye scatterers suspension, but the polymer film is in a dynamic vacuum to prevent photodegradation. Since the polymer is a solid medium, one location on the film produces the same reproducible spectrum. To observe the narrow emission spikes, the trick is to look at the small excitation area. Fig. 3 shows the spectra of three different locations on the polymer film. The excitation area is approximately 1 mm by 100 μm. The inset is the same polymer film but the excitation area is 6 mm by 100 μm. We observe that the narrow peaks are apparently lost.

There is a parallel between the number of pulses for the dye scatterer suspension and excited area with polymers. Observation of narrow emission lines occurs with a small value for each. Larger amounts lead to a recorded emission with apparently smoother emission curves. A small area in the polymer film corresponds to few excitation pulses in the dye scatterer suspension. The smooth curves result from the superposition of many narrow emission lines.

It is tempting to call the very narrow lines laser emission. The best way to identify the nature of the emission is to use photon statistics [8]–[10]. During a short time, one counts the number of photons arriving at the detector. This measurement is repeated for many observations and the resulting counts are placed in a histogram. If the observation time is short enough, then coherent light has one type of photon distribution whereas incoherent light has another [8]. These distributions are given as follows:

$$p(n, m) = \frac{m^n}{(1 + m)^{1+n}} \text{ incoherent} \qquad (1)$$

$$p(n, m) = \frac{m^n \exp(-m)}{n!} \text{ coherent} \qquad (2)$$

where $p(n, m)$ is the probability of observing n photons with a mean number m. The incoherent distribution of (1) is a Bose–Einstein distribution function, whereas the coherent distribution of (2) is a Poisson function. The most probable value for the Poisson distribution is the mean value m, whereas the most probable value for the Bose–Einstein distribution is zero. A very narrow range in both time and wavelength is necessary since an incoherent distribution can give a Poisson distribution for a long observation time [8].

The experimental setup was altered for photon counting. A streak camera was inserted between the spectrometer and the CCD camera. The streak camera changes a signal in time to a signal in space. The input emision spectrum is now recorded on the CCD array as wavelength versus time. Fig. 4 shows the time resolved spectrum for a single excitation pulse on the dye-TiO_2 suspension. A narrow time interval from 62 to 100 ps and a wavelength window of 0.15 nm is used to produce a histogram of photon counts. The one centered at 560.6 nm is a "valley," the other at 560.2 nm is an emission peak. Fig. 5 shows the resulting histograms for two locations in the time resolved spectrum. Fig. 5(a) is for 560.6 nm, shown with a theoretical fit for incoherent light. Fig. 5(b) is for 560.2 nm, shown with a fit for coherent light. The same set of data produces both distributions; this is a good check that the time used for the histograms is not too long. We thus conclude that the narrow emission lines are coherent radiation.

Similar photon counting experiments have been performed for the dye-TiO_2 polymer system [6], the π-conjugated polymer film [11], and the zinc-oxide nanoclusters [12]. The results are the same; the very narrow emission lines from the random systems are coherent radiation.

III. Fourier Transform

Since the narrow emission lines have been shown to be coherent, the natural question of feedback arises. This question can be answered with the help of the Fourier transform (FT).

The FT is often used to look at the frequency domain given time domain data. When the FT is applied to laser emission spectra, the fundamental resonator length is recovered [13].
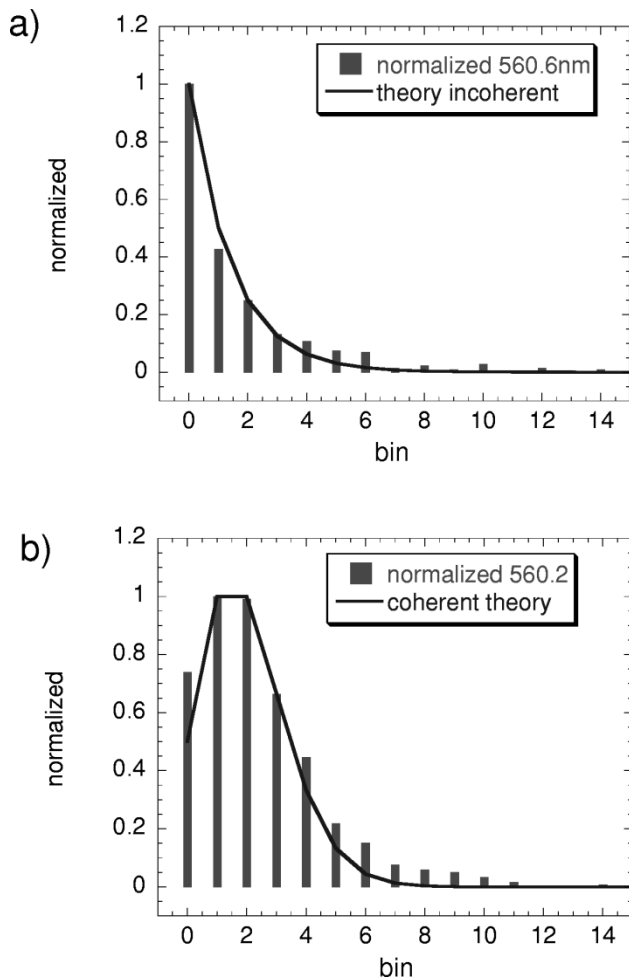
Fig. 5. Photon counting for two regions in Fig. 4. (a) A "valley" centered at 560.6 nm. (b) A "peak" centered near 560.2 nm. The lines through the data are fits using incoherent disrtibution of (1) and coherent distribution (2), respectively.
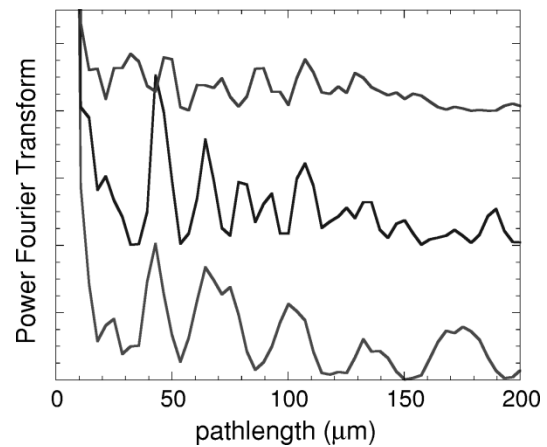

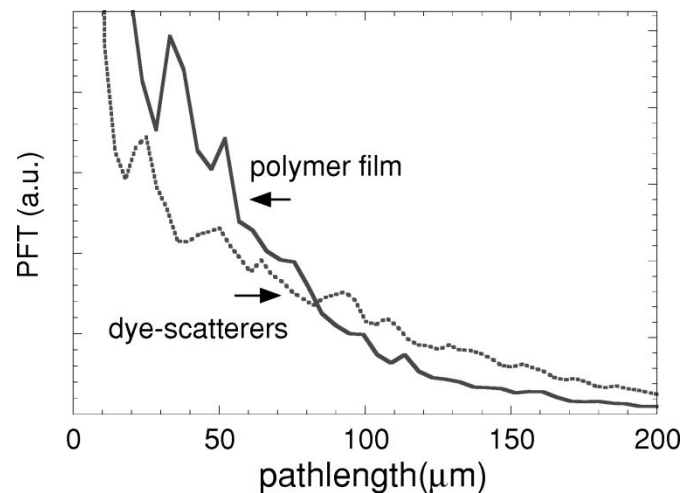Fig. 6. PFT spectra of the three-emission spectra shown in Fig. 1.

Fig. 7. Average power fourier transform spectra of 100 random laser emission spectra of dye-TiO_2 suspension (broken line) and the π-conjugated polymer (full line).

The units of the emission spectrum need to be in wavenumber (1/length) so the units in the FT are in length. The expected FT consists of equally spaced harmonics with decreasing amplitude [13]. The actual separation of harmonics for a Fabry–Perot resonator is nL/π where n is the index of refraction and L is the mirror separation. The FT procedure works for a ring configuration as well [14], [15]. For the ring, the mirror separation is replaced by half a circumference and the resulting FT harmonics appear at multiples of $nD/2$, where D is the ring diameter.

Fig. 6 is the power Fourier transform (PFT) of the three spectra of the dye-TiO_2 suspension from Fig. 2. All of the PFT spectra have many peaks, but none seem to have a sequence of regularly spaced peaks. The large components near zero correspond to a "dc-offset, " meaning that the emission spectrum has only positive values.

A remarkable thing happens when the many individual PFT are averaged. If the *laser emission* spectra are averaged for 100 excitation pulses, then the resulting spectra is the smoooth spec-turm seen in the inset of Fig. 2. On the contrary, if the average of 100 separate PFT are averaged, then one gets Fig. 7. On the average, PFT harmonics are *more regular* than for any indi-

vidual PFT. In fact, four harmonics are easily visible in Fig. 7. The PFT is complicated by the fact that there are two cavities, one at $nL/\pi = 24.5$ μm and the other at 31 μm. Taking the index of refraction to be that of methanol, namely $n = 1.329$, a Fabry–Perot cavity would have a distance of 58 or 73 μm, respectively, between the mirrors. If the resonance cavity is circular, then the microring type resonators have diameters of 37 and 47 μm, respectively.

The same process of averaging PFTs of laser emission spectra was applied to the polymer film. Instead of different pulses, the PFT were averaged over different illuminated 1 by 100-μm areas. One fundamental cavity length of 16.5 μm emerged from the average PFT [16]. The prominent peaks in the curve of the PFT in Fig. 7 are the second and third harmonics. With a index of refraction of 1.8 for the polymer, a Fabry–Perot mirror sep-aration of 29 μm or a circular cavity with diameter of 18 μm could be calculated. The polymer film has been imaged and bright spots with diameters less than 50 μm have been observed [16]. The size of the bright spots corresponds to the dominant cavity in the average PFT.

The very regular harmonics in the averaged PFT indicates a strong selection process for random lasers that selects a pre-

ferred resonator length within the gain medium. The following plausibility argument was proposed to explain the selection process [16]. For any laser, the gain must exceed the total loss to achieve lasing. For a disordered gain medium the total gain is proportional to the length traveled. There is a critical length L_γ, where lasing begins. We assume that the probability of traveling back to the start of a random walk through the scatterers and gain media after traveling a length L decreases exponentially: $[\exp(-L/l^*)]$, where l^* is the mean-free path in the medium [16]. Closed loops are necessary to achieve coherent emission. Any closed loop with $L < L_\gamma$ will not lase.

There are strong cutoffs on both sides of L_γ. On the small side, round trips with these lengths have insufficient gain to lase. On the large side, there are paths with higher gain, but exponentially fewer in number. The pathlengths that lase are selected to be very near L_γ, and therefore they are nearly *identical*. This is one of the main properties of random lasers.

The dye-TiO_2 suspension shows two fundamental lengths that may contradict the above explanation. The polymer film thickness is about 1 μm and the emission wavelength is about 0.6 μ for the illuminated area of 1 by 100 μm; thus this is almost a two-dimensional system. In this system, there is a single dominant resonator length. On the contrary, the dye-TiO_2 system is a liquid and is thus three-dimensional. Layers of liquid closer to the surface have a greater excitation since they very efficiently absorb the excitation and leave less for deeper layers. The gain in directions parallel to the surface and perpendicular to it is likely anisotropic. It may be that one cavity length is parallel to the surface of the cuvette and the other is perpendicular, with each direction having a different gain coefficient. It is also possible that deeper suspension layers with different gain give rise to different resonators.

In summary, the coherent narrow emission from random systems can justifiable be called random lasing. Feedback occurs for closed loops with a round trip length within a narrow range of the minimum length required to achieve lasing.

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