

Review of Open Cavity Random Lasers as Laser-Based Sensors

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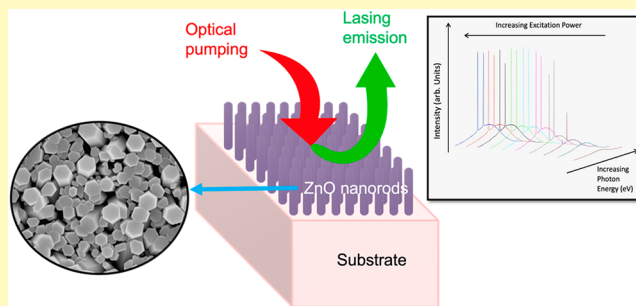
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ABSTRACT: In this review, the concept of open cavity lasing for ultrasensitive sensing is explored, specifically in driving important innovations as laser-based biosensors—a field mostly dominated by fluorescence-based sensing. Laser-based sensing exhibits higher signal amplification and lower signal-to-noise ratio due to narrow emission lines as well as high sensitivity due to nonlinear components. The versatility of open cavity random lasers for probing analytes directly which is ultrasensitive to small changes in chemical composition and temperature fluctuations paves the path of utilizing narrow emission lines for advanced sensing. The concept of random lasing is first explained followed by a comparison of the different lasing threshold that has been reported. This is followed by a survey of reports on laser-based sensing and more specifically as biosensors. Finally, a perspective on the way forward for open cavity laser-based sensing is put forth.

KEYWORDS: random laser, laser-based sensing, biosensor, cancer detection, Huntington's disease detection, brain tissue detection, flexible laser, fiber random laser



The key challenges in chemical and biosensing relate to measuring the target molecule with maximum-signal-to-noise ratio, highest specificity, and lowest limit of detection.¹ A random laser, that relies on an open cavity system, has the advantage in probing analytes and biological substances directly while maintaining its lasing characteristics. Hence, it is a promising alternative for biomarkers, biosensing, and biocompatible sensors.^{2–4} For example, the sensing capabilities from random lasers, which potentially could be superior to other optically based sensors already available, arises from the capability of achieving high signal amplification and high sensitivity due to narrow emission lines which are always favorable over broadband emitters. For a random laser to be adopted into any existing technology, the lasing threshold must be small, which is at present still a challenge for random lasers. As such, significant progress has been made over the years to further improve random lasers by improving the design and attributes of a random laser design that significantly lowers the lasing threshold, enhance the lasing emission and lasing intensity as well as providing lasing tunability elements that may be of paramount importance for ultrasensitive sensing.^{5–7} A review of current threshold values obtained for various materials is presented in the section [Improvements in Lasing Threshold](#). This includes fundamental investigations into improving control over the random lasing threshold and scattering characteristics that can bring new opportunities to utilize such open cavity random laser systems for a range of

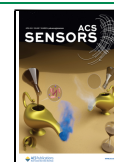
applications such as cancer detection, photodynamic therapy, document encoding, displays, speckle-free imaging, microscopy, and sensing applications.^{8–12}

The current state of laser-based optical biosensors has mostly evolved around the use of laser dyes as a gain medium.^{13–16} Such dye-based sensors typically result in a broad lasing spectrum (10 to 30 nm line width—which is comparable to fluorescence-based sensors) and are subject to degradation of the laser dye. As such, solid-state gain medium for random lasers has been extensively investigated by researchers to replace the need for laser dyes to achieve lasing.^{17–21} Certain types of random lasers employing a solid-state gain medium with high scattering provided by the gain medium itself and have been shown to provide a narrow emission line.^{22–24} Hence, degradation issues could be addressed by employing a solid-state gain medium with current laser-based biosensors. A review of random laser-based biosensors is provided in detail in the section [Random Laser Biosensors](#). Finally, this paper concludes with a way

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forward for random lasers with solid-state gain medium as biocompatible flexible laser-based sensors.

■ CONCEPT OF RANDOM LASER

A random laser relies on an open cavity system whereby no physical set of mirrors like a conventional laser system is required to provide feedback. The optical feedback from a random laser is based on the trapping of light inside the gain media (that can be easily achieved by utilizing nanostructures) which results in amplified stimulated emission into the scattering modes for lasing to occur. It was first explored in 1966 by a group led by Nobel laureate N. G. Basov where one of the cavity mirrors was replaced with a scattering medium and lasing emission was observed.²⁵ In this 1966 experiment, one of the mirrors in the Fabry–Perot cavity was replaced with a scatterer, and lasing was observed—the first evidence of strong scattering that can produce laser emission.

This work led to Letokhov modifying the diffusion theory to include gain, in order to understand the photon's random walk in a high scattering region.²⁶ In a simple model description, the lasing in a random laser can be described by the set of rate equations for the excited-state and photon populations:

$$\begin{aligned}\frac{dN_1(t)}{dt} &= P(t) - \frac{\beta q(t)N_1(t)}{\tau} - \frac{N_1(t)}{\tau} \\ \frac{dq(t)}{dt} &= \frac{\beta N_1(t)}{\tau} [q(t) + 1] - \frac{q(t)}{\tau_c}\end{aligned}\quad (1)$$

where $N_1(t)$ is the number of excited molecules, $P(t)$ the pump rate, $q(t)$ the number of photons in laser modes, τ the spontaneous emission lifetime of the dye, and β the β -factor for a random laser that involves the average numbers of lasing modes. The cavity-decay time, τ_c , depends on the transport of light in the medium, which in the diffusion approximation is given by the diffuse transport time $\tau_c = \frac{L^2}{8D(\lambda)}$, where $D(\lambda)$ denotes the diffusion constant of light and L is the size of the multiple scattering objects (assuming equal size in all dimensions). The diffusion constant is a dynamic transport property which is related to the static transport mean free path $l_t(\lambda)$ by the relationship $D(\lambda) = \sqrt{\nu} l_t/3$, with ν the transport (energy) velocity. By modifying $D(\lambda)$ and $l_t(\lambda)$, the threshold and laser process can be controlled.

Experimentally, Migus and Markushev found that grinding a crystal into fine powder offers optical gain through stimulated emission and multiple scattering.^{27,28} Shortly afterward, lasing from highly scattering medium was realized in 1994 when Lawandy found a way to generate a multimode laser spectra's temporal and spectral properties without an external cavity or known as random laser.^{29,30} This was followed by seminal work by Wiersma and Lagendijk^{31,32} and by Cao and co-workers on random lasing in ZnO and GaN powders,^{33,34} which set the scene for several detailed studies on different aspects of localized random lasing modes versus extended modes.^{35–37} The effort in understanding such random lasers was further expanded by theoretical studies addressing mode competition and lasing dynamics.^{38,39} Hence, with such versatility random lasers can be tailored to suit specific application needs.

Based on eq 1, clearly random lasers require the same conditions as conventional lasers to achieve the lasing condition—a gain medium that provides optical gain through

stimulated emission and partial trapping of light providing feedback. Light trapping in the case of random lasers is achieved through a multiple photon scattering process—propagation known as a random walk with the characteristic length scale as the transport mean free path (l_t). This kind of scattering is observed in Nature, for example, in butterflies, moths, and beetles.⁴⁰ Random lasers have also been inspired by the structure of butterfly wings.⁴¹ Figure 1 shows this random walk and bioinspired photonics from butterfly wings.

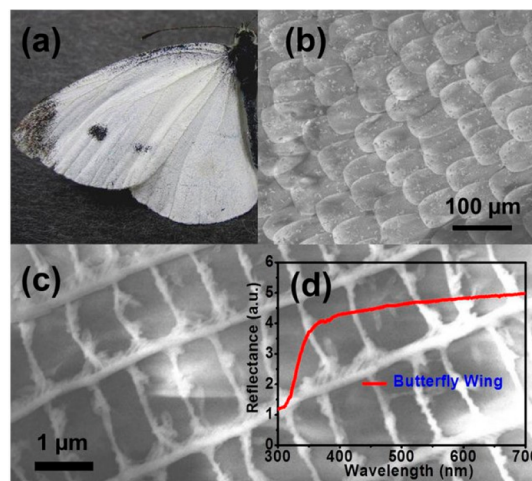


Figure 1. (a) Picture of a *Pieris canidia* butterfly; (b) low magnification scanning electron microscope (SEM) image of the wing scales; (c) high magnification SEM image of the wing scales; (d) reflectance spectrum of butterfly wing.⁵⁶

In some random lasers, the gain medium and scatterers are from the same material known as active material. Zinc oxide (ZnO) is the most common active material that has been investigated due to its high refractive index, high binding energy in room temperature (60 meV), and stability. A random laser using an active material provides several advantages such as simple experiments—only one material is required and the design can be focused on just the scatterers (e.g., population density, filling factor, thickness). The fabrication does not require a different gain material, and since such material is generally solid, the issue of concentration control can be eliminated. The only drawback is that refractive index must be high so that scattering would overcome absorption losses.⁴² This effect was explicitly studied in gallium arsenide (GaAs) powders whereby alumina particles were mixed with GaAs powder to increase the scattering probability for random lasing to occur.¹⁹ Based on the results, the authors proposed that lasing was not achieved in GaAs powder alone, as photons participated in the absorption much more than the scattering. Because of this, some random lasers are prepared with a separate gain material with the scatterers, where rhodamine dye is commonly used, even though the use of dyes comes with challenges of controlling concentration and degradation.^{43–45}

The gain medium of a random laser determines the emission wavelength. For rhodamine, the emission is between 550 and 610 nm. For ZnO random lasers, the emission is at ~380 nm, and for gallium nitride (GaN) the emission is ~365 nm.^{46,47} Small changes in the emission wavelength are likely to occur in semiconductor nanostructures due to minor changes in the bandgap from small fluctuations of the size of nanostructures, mostly observed from ZnO.^{48–50} Figure 2 shows the different

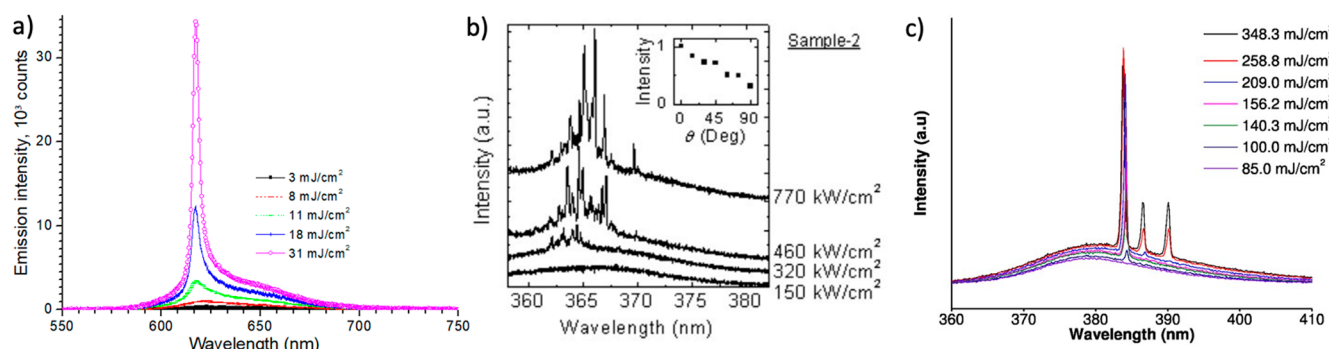


Figure 2. Emission spectra of (a) rhodamine dye mixed with TiO_2 nanoparticle at several pump powers with a concentration of $2 \times 10^{11} \text{ cm}^{-3}$, (b) GaN nanocolumns, and (c) ZnO nanorods.^{20,49,63}

random lasing emission wavelengths observed from several materials upon achieving the threshold. As observed from the figure, random lasers prepared by ZnO and GaN (as well as most solid-state semiconductor random lasers) show multiple emissions. This creates a problem for utilizing random lasers as laser-based sensors. Current random lasers have been engineered toward reducing the number of lasing modes. Methods include introducing point defects, controlling absorption, and altering the Mie resonances.^{50–62}

To achieve open cavity lasing, it is also important to trap the wavelength of light that can achieve gain. Early discussion of this phenomenon has often highlighted the close relationship to strong multiple scattering of light in disordered media, also known as photon localization or Anderson localization. However, over years of study it was found that this type of lasing was found in a very wide range of systems of different scattering strength, far beyond the regime where localization conditions could possibly play a role.¹² Different particle sizes looking into the effect of scattering in random lasing emission have been investigated with varying gain mediums.^{64–69} Further investigations yielded the relation of random lasing with photon random walk that defines the transport mean free path of these photons—a parameter to understand how far photons can travel in a highly scattered medium of a random laser that has gain.^{70–72} Below, we will briefly discuss some of the basic principles of light scattering of importance to understand the behavior of open cavity random lasers.

When light passes through particles or obstacles such as dust, mists, and powder, it causes the light to move in all directions. Rayleigh's Law as in eq 2 states that the intensity of scattered light is inversely proportional to the fourth power of wavelength.

$$I_s \propto \frac{1}{\lambda^4} \quad (2)$$

I_s refers to intensity of scattered light and λ refers to the respective wavelength.

There are two types of scattering: elastic scattering and inelastic scattering. Elastic scattering occurs when the direction of light propagation is changed, and the photon's energy is conserved. However, for inelastic scattering, the direction of light propagation and the energy of the photon both changes. The elastic scattering can be characterized using Rayleigh scattering, Mie scattering, and geometric optics or ray optics, which depend on the size of nanoparticles.

Rayleigh scattering refers to scattering of particles when the diameter is much smaller than the wavelength of scattered light

($d \ll \lambda$). The Rayleigh scattering's equation is crucial to determine scattering cross section, polarization of dielectric sphere, and radiation from a driven oscillating dipole. Inversely, geometrical optics are cases where the particle's radius is larger than the wavelength of light ($d \gg \lambda$). This is when light is treated as rays as they encounter big objects or optical systems generally 1000 times larger than the wavelength of light. On the other hand, Mie scattering refers to scattering from spherical nanoparticles that are almost the same size as the wavelength of light ($d \sim \lambda$). The idea of Mie scattering is to expand the incident plane wave in the Fourier series using appropriate basis functions that satisfy Maxwell's equation in spherical coordinates. The derivation of the Mie scattering leads to the determination of the differential and total scattering cross sections. For random lasing, scattering must be within the Mie scattering regime for Anderson localization of photons to work.⁷³ The Mie scattering cross section is shown in eq 3

$$\sigma_{\text{Mie}} = \left(\frac{2\pi}{k_{\text{med}}^2} \right) \sum_{n=1}^{\infty} (2n+1)(|a_n|^2 + |b_n|^2) \quad (3)$$

where $k_{\text{med}} = \frac{2\pi n_{\text{med}}}{\lambda_0}$. The coefficients a_n and b_n are given by

$$a_n = \frac{\mu m^2 j_n(mx) [x j_n(x)]' - \mu j_n(x) [mx j_n(mx)]'}{\mu m^2 j_n(mx) [x h_n^{(1)}(x)]' - \mu_1 h_n^{(1)}(x) [mx j_n(mx)]'}$$

$$b_n = \frac{\mu_1 j_n(mx) [x j_n(x)]' - \mu j_n(x) [mx j_n(mx)]'}{\mu_1 j_n(mx) [x h_n^{(1)}(x)]' - \mu h_n^{(1)}(x) [mx j_n(mx)]'}$$

where the j_n 's are the spherical Bessel functions of the first kind, h_n 's are spherical Hankel functions, and μ_1 and μ_2 are the magnetic permeability of the sphere and surrounding medium, respectively.

In random nanostructures, light waves are scattered by mesoscopic variations of refractive indices to create a complex wavelength dependent grainy mode structure ("speckle") within a certain volume of material. Both scattering mean path and transport mean free path are essential to describe the scattering process. The scattering mean path is defined as the average distance a molecule travels between collisions. Meanwhile, transport mean path is the average distance a wave travels before its direction of propagation is randomized. The relationship between scattering mean path and transport mean free path is given as eq 4⁷²

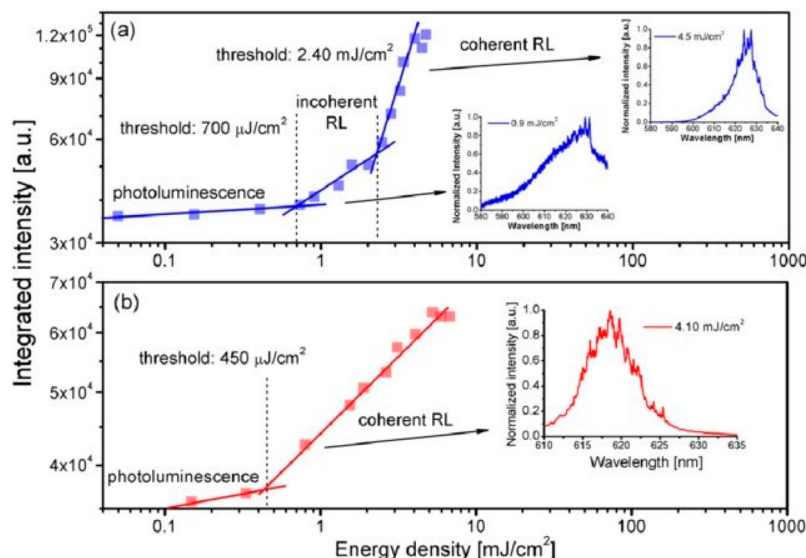


Figure 3. Integrated intensity dependences on energy density of pumping light for DCM doped PVK polymer layer before (a) and after (b) the rubbing process that explains the laser emission due to coherent and incoherent feedback. Insets contain averaged random lasing spectra observed at selected regions.⁷⁸

$$l_t = \frac{l_s}{1 - \langle \cos \theta \rangle} \quad (4)$$

Here $\langle \cos \theta \rangle$ is the average cosine of the scattering angle obtained from the differential scattering cross-section. Rayleigh scattering is an example of $\langle \cos \theta \rangle = 0$ or $l_t = l_s$, while Mie scattering may have $\langle \cos \theta \rangle \approx 0.5$, or $l_t \approx 2l_s$.

For an open cavity system, such as a random laser, the threshold would be high compared to traditional lasers as the system is very lossy.⁷⁴ Several factors affect the lasing threshold of a random laser, and the main factor is the scattering strength. Hence, many efforts have been focused on increasing the scattering strength which is determined by l_t . A quantitative analysis was carried out in which several random lasers were compared with their different l_t values that determines the threshold for the random lasers.⁷⁵ In the weak scattering regime, l_t is roughly the sample thickness, and the role of scattering is only to redistribute the amplified spontaneous emission. Most reported random lasers have l_t much smaller than the sample size but larger than the wavelength. The scattering in this case increases the residence time of light in the gain region owing to the multiple scattering.⁷⁶ In the strong scattering regime, l_t becomes equal to or smaller than the wavelength. Then, the recurring scattering event becomes important. This was observed in semiconductor powder, and the observed discrete lasing modes were argued to arise from ring cavities in the scattering medium formed by the recurrent scattering events.⁶⁵

■ INCOHERENT AND COHERENT RANDOM LASERS

There are two types of random laser, namely, laser with incoherent feedback and laser with coherent feedback. The incoherent random laser is the result of amplified spontaneous emission taking place inside a diffuse scattering volume and can be considered as the solution of an incoherent photon diffusion model with gain. Two characteristic length scales must be considered to identify incoherent feedback, i.e., gain length and average path length. The gain length can be defined as the distance a photon travels before generating other

photons. The average path length is the average path length of a photon that travels in the gain medium. Theoretically, the gain length and average path length are mutually dependent. As the amount of scattering increases, the average path length increases. When the average path is equal to the gain length, the number of photons increases.

In the late 1990s, the first batch of random lasers with coherent feedback was realized with disordered semiconductors and organic materials.²⁹ The coherent feedback mechanism is related to gain provided by specific scattering eigenmodes of the system. These eigenmodes could be potentially closed-loop paths where light is cycled several times like in a cavity, or alternatively these can consist of a subset of nonrecurrent, open pseudo modes of the scattering medium with sufficiently long dwell times and gain length.³² Coherent random lasing gives rise to narrow spectral peaks above the emission background when the lasing threshold occurs.⁷⁷ Early experiments discussed that coherent feedback in both strong and weak scattering regimes may be provided by recurrent scattering. However, the transport mean free path of photons must be considered to produce sufficiently strong scattering for a significant number of recurring paths to occur—it must be shorter than their reciprocal wave vector, known as the Ioffe-Regel criterion.⁷² This regime produces strong photon localization equivalent to Anderson localization. The localization regime is however much easier to achieve in lower dimensional systems such as 1D or 2D waveguides, and therefore open cavity lasers in these lower dimensions could particularly benefit from localization effects. For such closed-loop paths, the coherent feedback enables spatial resonance. Then, the spontaneously emitted photons are amplified through stimulated emission with mode structures derived from the spatial resonance.

Figure 3 shows the different emissions obtained in an incoherent random laser and a coherent random laser from the DCM-doped PVK polymer layer. Based on Figure 3a, Sznitko et al.⁷⁸ describe the emission from an incoherent random laser as starting by a broad emission (spontaneous photons) induced by photoluminescence whereby the emission peak

Table 1. Summary of Random Lasing Threshold in Different Random Lasers Where D Is the Scatter Diameter, CS Is the Coronal Section of Brain Tissues, L Is the Length of the Scatterer, T Is Thickness of Scatterer, and n Is the Refractive Index of the Medium

year	threshold value	gain material	scatterer	size of scatterer	ref
2021	23 kW/cm ²	Rhodamine 6G	Silver nanoparticles	D : 70 nm L : 0.7–1.2 μ m	85
2020	43.3 MW/cm ²	Rhodamine 6G	Polydimethylsiloxane	D : 17 μ m	86
2020	0.07 W/cm ²	Aluminum doped zinc oxide nanorods	Aluminum doped zinc oxide nanorods	D : 34 nm	54
2020	5 kW/cm ²	Zinc cadmium selenide/Zinc sulfide quantum dot	polymer-dispersed liquid crystals	10–15 nm	87
2020	160 kW/cm ²	4-(Dicyanomethylene)-2-tert-butyl-6(1,1,7,7 tetramethyljulolidyl-9-enyl)-4H-pyran	poly(dimethylsiloxane)	6 μ m	88
2019	318 kW/cm ²	Rhodamine 6G	Brain tissues	CS: 100 μ m	89
2019	2.75 MW/cm ²	Rhodamine 640	Titanium dioxide	D : 200 nm	69
2019	30.5 kW/cm ²	Luminescence polymer	Silver nanoparticles	100–300 nm	90
2018	80 W	Pyromethene 597 dye	Nematic liquid crystals	n.p.	91
2018	0.63 W/cm ²	Rhodamine B	Silk fibroin	D : 0.82–0.88 μ m	92
2017	267 kW	Rhodamine B	Hexagonal Boron nitride	D : 388 nm	67
2016	1.3 MW/cm ²	Rhodamine 6G	Photonic glass	D : 1.3 μ m	93
2015	7.75 MW/cm ²	Rhodamine 640	Gold nanoparticles	D : 20 nm	4
2014	9.5 MW/cm ²	Rhodamine 6G	Aluminum particles	D : 150 nm	94
2013	4.5 MW/cm ²	Rhodamine 6G	Latex nanoparticles	D : 51–380 nm	95
2010	0.5 kW/cm ²	Rhodamine 800	Bone tissues	T : 200 μ m	96
2009	600 kW/cm ²	Zinc selenide	Zinc selenide	several hundred nm	97
2004	198 kW/cm ²	Gallium nitride	Gallium nitride	D : 80 nm	98

increases linearly with increasing pump power. Then, above the energy density value of 700 μ J/cm², the integrated intensity grows rapidly, which confirms the onset of lasing. However, above the energy density of 2.4 mJ/cm², sharp spikes are observed in the emission spectra. The narrow spectrum confirms coherent lasing. However, coherent random lasers as shown in Figure 3b indicate a clear threshold at around 450 μ J/cm² whereby the intensity increases exponentially with pump power. As observed, the threshold of coherent random lasers is lower by almost half (450 μ J/cm²) compared to incoherent random lasers (700 μ J/cm²). This condition of double threshold behavior has also been observed in resonance-controlled ZnO random lasers.⁴⁸ Lasing at the first threshold appeared around exciton recombination energies, and the lasing threshold carrier density was comparable to the Mott density, whereas the lasing at the second threshold was induced by population inversion accompanied by electron–hole plasma recombination like conventional ZnO random lasers. However, for laser-based sensing, coherent random lasers should be considered, as fluctuation in the threshold can change the sensing parameters accordingly.

IMPROVEMENTS IN LASING THRESHOLD

An important factor for laser-based sensing, other than controlling the lasing emission, is achieving a low lasing threshold. If the photons participate in the scattering event much more than in other events such as absorption or just pass through the sample, then random lasing is easily achieved, and hence the threshold would be low. Reducing the threshold is critical not only for lowering the operation costs but also for integrating as an electronically pumped laser. The current method of making random lasers rely on optical pumping, however, increased efforts have been pushed toward achieving

stable electrically pumped random lasers, since the threshold for achieving it has gone down significantly.^{79–81}

Table 1 summarizes the thresholds from random lasers prepared from different scatterers and gain mediums. Additional factors such as concentration, size, and population density of scatters, concentration of gain, and choices of material (as each material has different refractive index) play a role to determine the threshold of a random laser. It is evident from the literature that the threshold is low when the gain materials are semiconductors. The gain material can be tailored to nanoscale sizes that are within the Mie resonances for random lasing to occur. Increasing the scattering strength and exploiting the resonances in scattering coefficients (Mie resonances) with surface plasmon resonances are other ways of reducing the random lasing threshold.^{82–84}

To cater for high-precision sensors (especially biosensors) that require wavelength tunability, random lasers can be designed to have tunability in the emission wavelength. This contrasts with conventional lasers whereby the emission wavelength is fixed due to a fixed cavity, and tuning can only be done at either double or triple the frequency using a frequency double placed at the laser head. In a random laser, however, fine-tuning of the wavelength is achieved by manipulating the gain medium or the scattering elements. Tunability can be achieved by changing the weight fraction,⁹⁹ particle size,⁵⁹ and photonic crystal lattice¹⁰⁰ and adding a nonfluorescent dye to the gain medium.⁵³ A more recent approach to tuning a random laser is by doping the gain material with metal or inorganic elements that shifts the bandgap of the material.^{6,54,101} The threshold for these doped random lasers varies but is much smaller (at least 1 order of magnitude) than the material without doping elements.

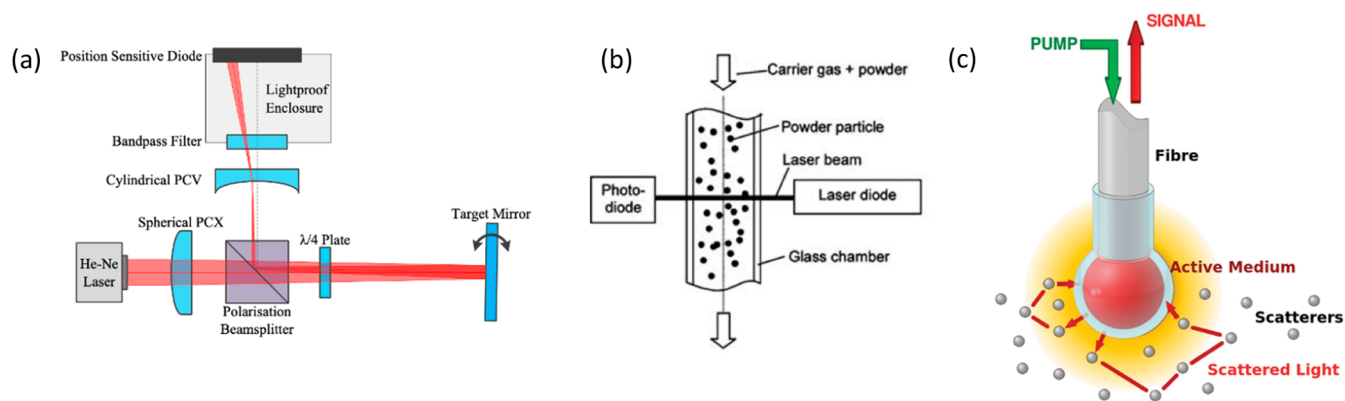


Figure 4. (a) Schematic diagram of the laser-interferometry-based sensing for misalignment detection. (b) Powder delivery rate sensor. (c) Random laser based optical sensor.^{104,113,114}

LASER-BASED SENSING OVERVIEW

Laser-based sensors have been reported to address critical challenges in creating reliable, real-time, and cost-effective monitoring integrated systems which are robust to changes in chemical composition over extended periods of time.¹⁰² The sensors must be precise to make them efficient especially for monitoring greenhouse gases for near- and long-term applications in environmental monitoring, measuring, and tracking ultra-high-precision movement particularly as a form of detecting misalignment in high-precision systems,¹⁰³ as a delivery rate sensor for laser-based additive manufacturing,¹⁰⁴ as well as being integrated into a laser measurement system known as LMS for preventing accidents.¹⁰⁵

Common detection characteristics vary according to whether the application is macroscopic or microscopic. For the automation industry (and most macroscopic applications), for example, the control engineer and software developers rely on laser-based sensing to identify if a particular target or object has arrived at a particular location. The common characteristic is detecting changes in the amount of light reflected from the object using sensors that are diffusive, reflective, or through-beam. Lasers are generally used for sensing further than 50 m. They can also be used to detect and track moving objects.¹⁰⁶ For microscopic applications such as for detection of gas molecules, the characteristics investigated include side-mode suppression ratio, tuning range, spectral emission line width, frequency stability, and wavelength modulation.¹⁰⁷ Combined with fiber optics, lasers can also be used to detect ulcers in the intestines. The detection characteristics rely on the distinctive spectral differences in Raman spectra.¹⁰⁸ Lasers are also used to study the internal structure of microorganism and cells.¹⁰⁹ Remote sensing is another form of laser-based sensing with LIDAR (Light Detection and Ranging) being the most popular choice. The distance, height of objects from the ground, and chemicals in the air are measured by light reflection.¹¹⁰

Perturbation measurements for detecting explosives utilizes the amplified spontaneous emission (ASE) whereby changes in intensity at a range of emission wavelength are recorded with high sensitivity.¹¹¹ Laser-based sensing has also been used to trace hazardous gas detection.¹¹² The laser is combined with a photoacoustic effect whereby light pulses are used to detect small concentrations of trace gases. Thus, the sensor should be highly sensitive (by obtaining narrow line width), tunable in wavelength, stable, and reliable. Figure 4 shows some examples of laser-based sensing.

For the detection of misalignment in high-precision systems, laser-interferometry-based sensing has been used. The detection of misalignment of position and orientation measurement beam such as robot manipulators is shown in Figure 4a). For laser-based additive manufacturing, an optoelectronic sensor is developed to sense the metal powder delivery rate in real-time. The sensor consists of a laser diode, a photo diode, and a glass window as shown in Figure 4b). The components are installed in such a way that the laser beam emitted from the laser diode passes through the powder stream flowing inside the glass chamber, and subsequently is received by the photo diode. The photo diode is characterized with a good linearity between the illumination energy received by the diode and the output current that is converted later to a voltage signal through a current signal pick-up circuit.

A laser-based vision sensor was also reported in welding, as part of an integrated system for structural health monitoring.¹¹⁵ Quality of the welding is checked by identifying the associated defects. As it is extremely difficult to identify the quality of the welding by the human eye, a laser-based vision sensor provides a solution for controlling and maintaining the quality of the welding. A special type of welding; metal inert gas (MIG) welding, is an arc welding process in which a continuous solid wire electrode is fed through a welding gun and into a weld pool, joining the two base materials together.¹¹⁶ Commonly, the MIG welding is applied in robots to combine each part of the robots whereby a laser-based tracking sensor was utilized to achieve high accuracy of the MIG welding.

In surgery, random lasing has been used to differentiate tissue.¹¹⁷ According to the authors, different approaches have been investigated as a feedback mechanism during laser surgery which are mainly categorized as imaging and nonimaging techniques. However, imaging techniques have the disadvantage of being too slow to provide feedback before damage is done to the tissue, making nonimaging techniques more favorable. A random laser has the added potential to provide information on a volume of the tissue on the surface and can detect the tissue type within a microsecond which may stop an otherwise nerve damaging laser pulse. Moreover, a random laser has the advantage that it can be generated easily with ultrashort pulse lasers. Results reveal the possibility to use random lasing for tissue differentiation even in the case of thermal tissue damage. However, the lasing emission is very close to the lasing threshold in some cases. In this specific case,

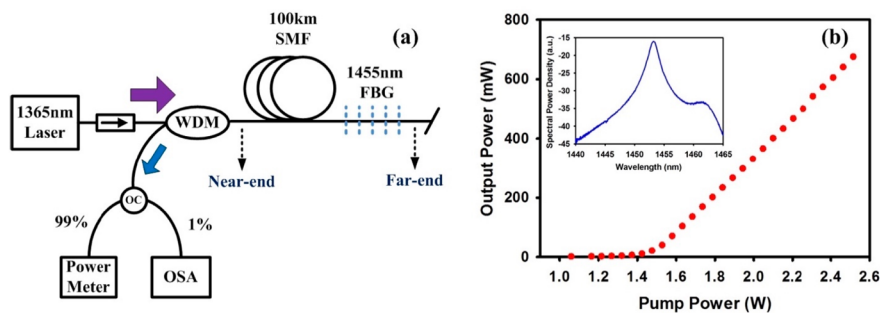


Figure 5. (a) Setup of the random fiber laser; (b) the output power as a function of the input pump power (inset: the output spectral shape corresponding to 2.3 W pump power).¹²⁹

R6G dye was used whereby the threshold tended to be high. A semiconductor gain medium for generating the random lasing is a potential alternative due to current reports on low lasing threshold (as shown in Table 1); however, this is yet to be investigated.

However, all the sensors mentioned above look into macroscopic elements or are observable by the naked eye. Open cavity laser-based sensing has the added advantage of analyzing analytes directly, which may not be the case with conventional laser systems that rely on a physical cavity. Therefore, random lasing is most suitable as optical sensors, which are largely dominated by fluorescence-based sensors, due to its simplified architecture. Optical sensor systems in several applications such as cellular temperature probe, fluorescence bioimaging, or anticancer therapies have been recently implemented.^{118–120} However, a comparison of a random lasing-based sensor with a conventional fluorescence sensor shows the sensitivity of random laser-based sensor exceeding 2 orders of magnitude.⁹³ The sensing mechanism was attributed to the modified gain and lasing threshold. No complex alignment was required, and the sensitivity was high. The mechanism of an open cavity laser-based sensor is shown in Figure 4c. Slight changes in scattering would change the threshold for lasing and shift the lasing wavelength. This makes it possible as a promising candidate for future biosensing and disease detection, which will be discussed in more detail in the following sections.

REMOTE SENSING

Lasers in general have been used for remote sensing applications. The most frequently used laser-based remote sensing application is in conjunction with LIDAR and DIALEX instrument.¹²¹ The DIALEX instrument is based on differential absorption spectroscopy and optical detection measured by the change in Doppler shift. This is commonly used to detect atmospheric species and natural target reflectivity.¹²² LIDAR on the other hand is based on the principle of backscattering whereby a light pulse is transmitted, and the backscattered light is detected and analyzed. The scattered light is detected earlier for closer objects compared to objects located much farther away. The return signal will not be the same length as the transmitted pulse but extended in time. For a more accurate measurement of the concentrations of atmospheric gases, especially gases at low concentrations, strong absorption features must be accessed. Since each molecular species or constituent has mid-infrared absorption characteristics, choosing the right laser source is also a crucial factor. Therefore, for assessing these characteristics, specific mid-infrared lasers has

been considered for remote sensing applications.¹²³ According to the author, among other vital considerations in choosing a laser source for specific remote sensing related to assessing molecular species is that (1) no cryogenic cooling is required to maintain lasing, (2) the beam is not divergent and astigmatic, (3) it uses high gain material (high power/energy output), and it has the advantages of (4) a simple design (for example, optical parametric oscillator (OPO) and difference frequency generation (DFG) have complex design), (5) low laser-induced damage, (6) narrow spectral bandwidth, and (7) broad tunability.

Apart from this, sensitivity may also be provided from scattering feedback. For example, a laser with external feedback is analogous to nonlinear Fabry–Perot resonators whereby the dynamic properties of injection lasers are significantly affected by external feedback.¹²⁴ The self-mixing effect has been applied for sensing applications whereby the interferometric system was built around the laser under optical feedback and results in many signals arising from laser feedback interferometry.¹²⁵ The signals detected by photodetectors can be easily collected and modulated, simplifying the structure and reducing disturbance noise and cost, as the laser itself can be used as the detector and does not need a reference beam, and the optical paths do not need additional optical components such as isolators, beam splitters, or target mirrors.¹²⁶ Another interesting innovation related to self-mixing interferometry is by integrating with random laser resonators that are operating at terahertz (THz) frequencies.¹²⁷ According to the authors, the emitted light reflects onto a surface with a random hole pattern, and by varying the external cavity length, the temporal dependence of the laser voltage is detected by a sequence of interference fringes that follow the bias-dependent spectral emission of the laser structure—a visible signature of the random laser sensitivity to the self-mixing effect. Results show possibilities of detector-less speckle-free nanoimaging and quantum sensing applications across the far-infrared.

Wideband remote sensing has been investigated using random fiber lasers.¹²⁸ Random fiber lasers have been used in combination with remote fiber sensors over ultra-long-distance fiber links. Very recently, the authors claim observation of instantaneous and linear responses to remote feedback disturbances using a Raman-gain-modulated power-balance model from the random fiber laser intensity, suitable for ultra-high-bandwidth sensing. On the other hand, random fiber lasers that do not have point reflectors have been shown to achieve thermal stability, which is beneficial for long-distance remote sensing using fiber optics.¹²⁹ This novel idea of utilizing such random lasers provides high-fidelity and long-

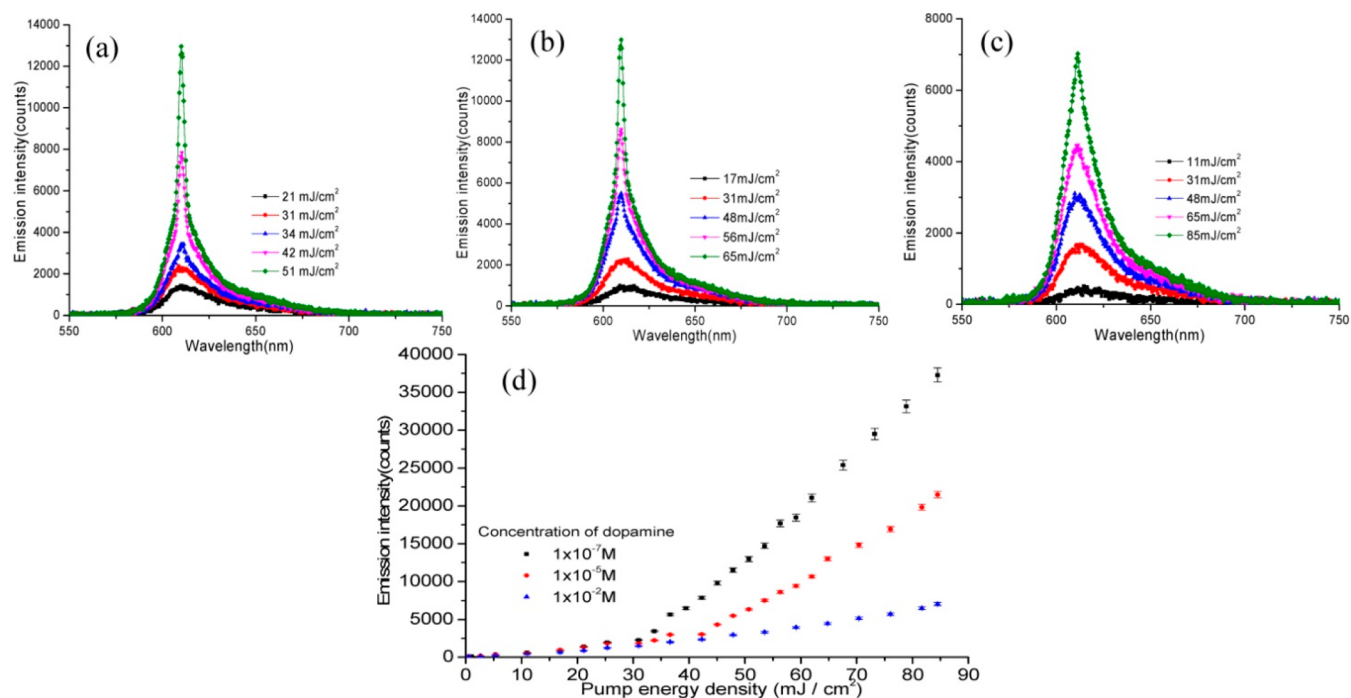


Figure 6. Emission spectra of Rh640/gold random lasers with copper(II) chloride (0.15 mM) with different concentrations of dopamine for different pump energy densities.⁴

distance transmission for the sensing signal. The authors claim a >20 dB optical signal-to-noise ratio (OSNR) over a 100 km distance. Intriguingly, the authors also found that the second-order random lasing emission has much better OSNR due to enhanced lasing efficiency by incorporating a 1455 nm fiber grating. The setup of such a device is shown in Figure 5.

RANDOM LASER BIOSENSORS

Random lasers have the added advantage of manipulating scattering to induce lasing. Therefore, changes in the scattering properties, for example, changes in cell structure, will change the lasing characteristics. As such, changes in biological analyte such as microorganism, cells, biomolecule, and other biological structure can correspondingly be detected.¹³⁰ This makes random lasers promising candidates for biosensor applications. The simplicity in its architecture is another compelling feature compared to other lasers whereby no physical cavity is required making them a very versatile type of laser well suited for such applications.

There are many types of biosensors such as piezoelectric, thermal, enzyme-based, tissue-based, and immunosensors which are used for different applications. In the food industry, biosensors can be used to detect bacteria and pathogens such as *Escherichia coli* (*E. coli*).¹³¹ One of the methods is by using redox potential measurements. When the analyzed solution reacts with the biosensor, it will immobilize on a particular semiconductor structure, and the response is recorded through changes in the modulated pulsed LED source, embedded within the system. A biosensor has also been developed for healthcare monitoring with the combination of microfluidic sampling, multiplexed biosensing, and transport systems. To improve the wearability and ease of operation, all components are integrated, miniaturized, and combined with flexible materials. These systems rely on fluorescence/spontaneous emission sources; hence the amount of bacteria/substance

required are high. However, to detect low concentrations or for early detection and diagnosis, laser-based sensing is required.

For laser-based biosensors, different types of lasers have been proposed and investigated. An intracellular microlaser is a good example whereby optical microresonators (small resonators) are used to confine light inside biological cells to measure internal stress and dynamic fluctuations at a sensitivity of 20 Pa, an order of magnitude better than direct image-based analysis.¹³² The authors noticed that cells under uniaxial stress give rise to splitting of the laser line, and small deformation of the cells supports laser oscillation. To excite and collect the light emitted from the tissues, an optical fiber was punctured into the tissue. No clear cavity was defined in the experiment; however, a gain medium was inserted in the cells, and excitation of the gain medium was done by optical pumping from an external source—a very similar principle to random lasing generation that utilizes an open cavity and relies on the scattering between the cells to confine the light for lasing to occur. However, based on the size of the droplet, authors claim the lasing mechanism to be due to whispering gallery modes. Whispering gallery mode lasing has also been utilized to observe contractility of cardiac cells.¹³³ Fluorescent polystyrene microspheres with diameters ranging from 10 and 20 μm were used as the gain medium. The authors showed that changes in cardiac contraction induce shifts in the laser emission wavelength. These investigations show the high sensitivity that laser-based sensing can achieve especially when it relates to microscopic biological events.

Conventional lasers with a Fabry–Perot cavity have also been investigated to explore cell parameters such as the size, shape, and refractive index. For instance, Song et al. discovered that the effective refractive index of a kidney cancer cell is 1.399 with 0.1% accuracy and an average cell size of 17.8 μm .¹³⁴ The experiment was carried out by capturing a single kidney cancer cell into the laser cavity, which resulted in a

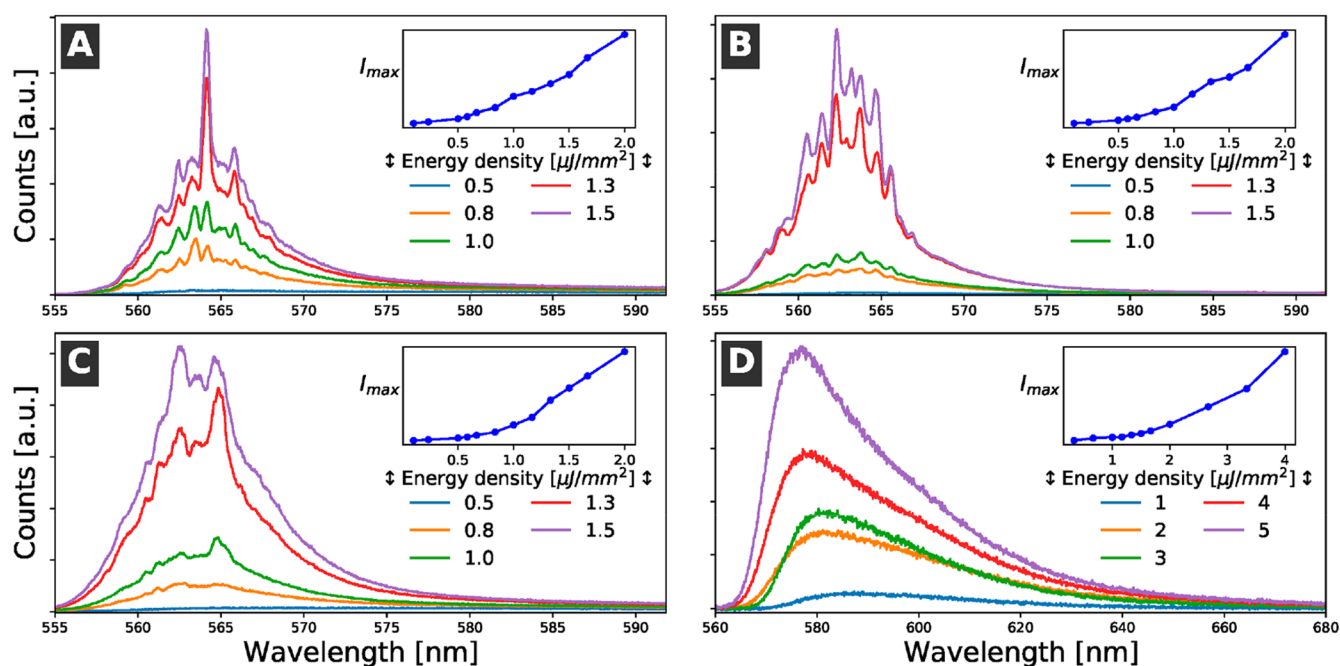


Figure 7. Evolution of the emission of the sample with increases in the pumping power density. The blueshift of the spectra, the emergence of narrow (fwhm <1 nm) lines, and the dramatic increase in emission intensity were all due to the coherent RL. (A) NT (nontransfected N2A cells). (B) HTT-Q23 (N2A cells transfected with the nonpathogenic pEGFP-Q23). (C) HTT-Q74 (N2A transfected with the pathogenic pEGFP-Q74). (D) Petri dish without cells. In this case, amplified spontaneous emission instead of random lasing was detected.¹⁴¹

spectrum shift. Recently, a similar principle was also applied to detect the concentration of cells in yeast with a sensitivity of 0.121 nm/(amo/mL) (2×10^{-7} nm/(cells/mL)) and a resolution of 3.65×10^5 cells/mL.¹³⁵ To improve the sensitivity, authors applied dielectrophoresis to make the final concentration of aggregation 50 times the initial state. Monitoring changes in refractive index in a Fabry–Perot cavity was also investigated in red and white blood cells.¹³⁶ The authors were able to distinguish the type of cells on a correlation coefficient threshold of 0.8. Identifying spectral properties of cancerous cells was also reported from investigating the mode confinement resulting from wavelength shifts.¹³⁷

For random lasers, utilizing them as biosensors is mostly due to the ability in achieving lasing with smaller gain material diameters compared to that of whispering gallery mode and conventional Fabry–Perot lasers. One particular example is to use random lasing to detect dopamine.⁴ A set of different dopamine concentrations were added to a solution consisting of gold nanoparticles, copper(II) chloride, and Rhodamine 640. In the detection process, dopamine triggers the aggregation of gold nanoparticles, where higher dopamine concentration can result in faster and larger aggregation of gold nanoparticles. The sensing parameters consist of the emission peak shift, emission peak line width, signal-to-noise ratio, and lasing threshold. The emission profile is shown in Figure 6. All these parameters contribute to just 250 SNR at 35 mJ/cm² of the lasing threshold. The detection limit of the system is $\sim 1 \times 10^{-7}$ M.

Another example is a plasmonic random laser that is used to detect Immunoglobulin (IgG) through specific binding to protein. The silver nanoparticles were self-assembled on a fiber facet by a polyvinylpyrrolidone (PVP) assisted reaction. The detection limit was as low as 0.68 nM.¹³⁸ There are also other plasmonic nanolasers that do not rely on scattering feedback

but rather on localized surface plasmons that are utilized as biosensors. Galanzha et al. have shown the capabilities of utilizing plasmonic nanolasers for biomedical imaging and a potential for photothermal disruption of cancer cells—a very novel approach in targeted cancer therapies.¹³⁹ The authors use gold nanoparticles surrounded by silica shell doped with a fluorescent dye as the gain medium. Plasmonic random lasers generally require longer pathways to achieve lasing and, in the past, require a high concentration of the gain medium in the cells, which limits the use of plasmonic random lasers in biomolecules and cells under investigation. However, current advancement in the gain medium used for random lasers, the low threshold that it can achieve, and very good detection limit as mentioned by Gather et al. show that plasmonic random lasers are very promising candidates for future biosensors.

RANDOM LASERS FOR DISEASE DETECTION

Random lasing has the advantage of sensitive detection for changes in biological cells. This discovery has led to using random lasers as detectors for diseases that involve changes in cells, most of them cancer related, for example, to study the marked PLCD1 gene therapy effect on human breast cancer. The different breast cancers give rise to either coherent or incoherent random lasers, hence making them unique identifiers of specific breast cancers.¹⁴⁰ For observing the actin cytoskeleton of AdHu5-PLCD1-infected cells, cells with or without AdHu5-PLCD1 were cultured on coverslips, soaked for 10 min in 4% paraformaldehyde, permeabilized in 0.1% Triton X-100 for 4 min, blocked with 1% bovine serum albumin for 1 h, and incubated for 1 h with rhodamine-conjugated phalloidin at room temperature. 4-(Dicyanomethylene)-2-tert-butyl-6-(1,1,7,7-tetramethyljulolidyl-9-enyl)-4H-pyran was selected as the gain media. The light emitted by the dyes is scattered in the biological tissues, which act as scatter centers. Coherent random lasing was associated with

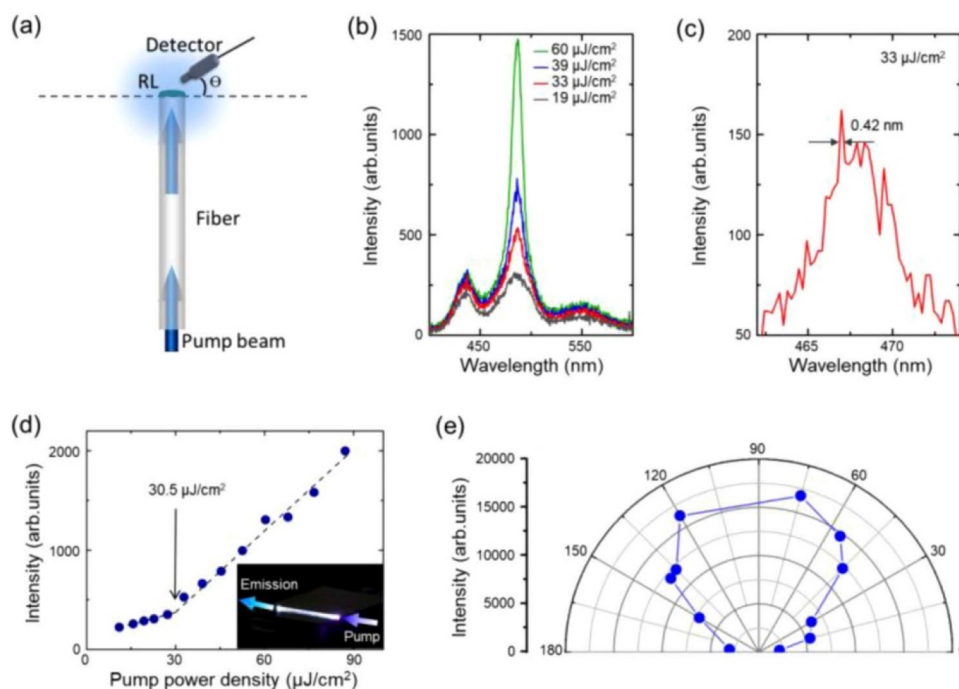


Figure 8. (a) Experimental setup of plasmonic random lasers on fiber facet. (b) Emission spectra of the plasmonic random system on fiber facet obtained under different pump power densities at a detection angle of 45° . (c) Specifics of emission spectra at pump power density of $33 \mu\text{J}/\text{cm}^2$, recorded by a high-resolution spectrometer with a resolution of 0.1 nm. (d) Emission intensity of the random lasing mode at 468 nm versus the pump energy density. Inset: corresponding images of the operating random laser on the fiber facet. (e) Integrated intensity of the random laser as a function of the detection angle.¹³⁸

the human breast cancer tissues that do not receive PLCD1 gene therapy, while incoherent random lasing was related to the tumor tissues that receive PLCD1 gene therapy. HE staining images attest to the fact that the distribution pattern of the breast tumor tissues that received PLCD1 therapy (AdHu5-PLCD1) exhibits less disorder, and the organizational structure of these tissues exhibits much less irregularity than that of the AdHu5-EGFP breast tumor tissues. This remarkably shows possible application of random laser in marking the process of gene therapy.

Most recently, random lasing has been utilized to detect mutant Huntingtin expression in cells related to Huntington's disease.¹⁴¹ According to the authors, the morphological/structural changes induced by mHTT expression can modify the scattering strength and the optical behavior of an environment when light propagates through it, resulting in differences in the random lasing signal compared to the signals arising from cells that express the wild-type *HTT* form. Random lasing emissions were obtained from transfected (EGFP-HTT-Q23 or EGFP-HTT-Q74) and untransfected cells resulting in a high-dimensional spectral data set. This data set was analyzed by using a multivariate statistical analysis based on principal component analysis and linear discriminant analysis. Lasing emission from this experiment is shown in Figure 7.

The high response at low concentration makes random lasers like these very promising candidates for biosensing and disease detection. However, the gain material utilized was fluorescent dyes which have problems related with dye degradation, self-absorption (can overcome by careful consideration of volume required), and the need to replace and dispose of the dyes for each measurement.¹⁴² To overcome these issues, semiconductors can be replaced as

the gain medium; however, no implementation of random lasers prepared with semiconductors (as scatterer and gain medium) as biosensors or disease detection and monitoring have been achieved to date—a very promising avenue especially for advancement in early detection of deadly diseases.

■ SENSITIVITY OF LASER-BASED SENSING

Increased interest in laser-based sensors comes from providing two main advantages: increased sensitivity and accuracy. The commonly used optical sensor is fluorescence-based and has low signal amplification due to the broad emission line, high signal-to-noise ratio, the absence of nonlinearity, and low sensitivity compared to the laser-based sensor. These weaknesses from fluorescence-based sensor can be solved using a random laser.

For a random laser as a pH sensor, various parameters are measured to identify the sensitivity of the sensor.⁹³ The sensitivity of a pH sensor is determined by the value of the relative absorption cross section, quantum efficiency, excited-state lifetimes, and gain cross section. The optimal sensing conditions and a maximal sensitivity 200 times higher compared to the fluorescence-based sensor was recorded by the authors.

Another example of sensitivity measurement from a random laser-based sensor is obtained from a random laser temperature sensor.¹⁴³ Its thermal sensitivity, S , was defined as $\frac{1}{R_0} \frac{dR}{dT}$, where R_0 is the intensity ratio at room temperature from the graph of the linear function intensity ratio of the signals against temperature. Thermal sensitivity up to 0.020°C have been recorded by the authors, which is at least 1 order of magnitude

larger sensitivity than that reported for fluorescence-based temperature sensors.¹⁴⁴

The brain is the most complex part of the human body. The brain has uneven surfaces which makes it a good candidate for random lasing if light can be scattered efficiently. Studies have shown that a random laser can detect changes in brain tissues' structure and composition.⁸⁹ Four different regions of the brain were investigated: the corpus callosum region, cortex, corpus striatum, and amygdala cortex. It was done by slicing the brain and soaking in different concentrations of Rhodamine 6G. The sensitivity was then measured by the emission spectra (at 565 nm), the value of full width at half-maximum of the spectrum, and the threshold value. Results show that the brain provides enough scattering for random lasing to occur. This work can be developed further by incorporating the advantage of a random laser as a speckle-free imaging technique whereby images of the brain can be improved.

CONCLUSION AND WAY FORWARD

In this review, the advantage of laser-based sensing is put forth as future sensors in the fields of precision sensing, remote sensing, and, mostly, biosensing. A comparison between different lasers and the potential of random lasers is highlighted with emphasis on the sensitivity elements as well as the detection of biological substance, diseases, and the changes in cell structure. The elements in laser-based sensors that make them superior to fluorescence-based is the narrow line width that gives rise to high signal-to-noise ratio and sensitivity of changes within the nanometer range. For random lasers, changes in the scattering of light changes the emission spectrum and, hence, small alteration of cell boundaries can be detected. In addition, detection of low concentrations of substances makes it advantageous for early detection of diseases which was proven for Alzheimer's, different types of cancer, and Huntington's disease.

Developing a random laser with low threshold pump power is one of the main challenges in utilizing random lasers as optical sensors. However, in realizing a random laser as laser-based sensors, flexibility is also key. Hence, some recent works have focused on flexible random lasers, electrically and optically pumped. One possibility is by constructing random lasers from optical fibers as fiber systems are well-established and readily viable. Several attempts in constructing random lasers from optical fibers have been made, involving the use of dye as the gain medium.^{14,138,145} Figure 8 shows one example of using optical fiber as a random laser. Authors report a red shift of the wavelength due to changes in refractive index caused by the binding of the protein.

A more practical approach would be to embed nanostructures that can provide gain, for example, ZnO, GaN, and GaAs. Implementing ZnO in fiber has successfully been demonstrated for other applications, mostly for sensing.^{146–149} Several detection mechanisms have been put forth. For instance, Bora et al. reported average coupling efficiency of ~2.65% that changes the sensitivity when exposed to different chemical vapors.¹⁴⁷ Kapoor et al. reported the use of bilayers of copper–zinc oxide in sensing the refractive index of liquids by fiberoptic SPR sensor.¹⁴⁸ Sensitivity increases with increasing zinc oxide layer thickness until 15 nm and then decreases above this value. On the other hand, last year Wu et al. proposed an optical fiber capped with a ZnO microwire whereby the tip acts as a Fabry–Perot cavity for monitoring deep UV light.¹⁴⁹ Changes in refractive index induced by

variations in the concentration of photogenerated carriers exhibits a red shift in the lasing spectrum with a sensitivity of 0.288 nm/(W·cm⁻²). However, refining according to the requirements of a random laser is yet to be explored. Generally, for random lasing the size of the nanorods is within the nanometer scale, much smaller than that used by Wu et al. However, the pumping source is the same for ZnO. Currently, utilizing GaN or GaAs on fiber for sensing has not been investigated, although random lasing from these gain materials shows promising results. The most used candidate is ZnO. Hence, it would be interesting to implement this on fiber and test its sensing capabilities which may be more sensitive than Fabry–Perot.

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Notes

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VOCABULARY

Random laser, A laser that relies on light trapping through multiple scattering process. The lasing characteristics such as onset of threshold before lasing occurs, narrow spectral width and coherent is the same as conventional lasers however the emission output directionality is random; Laser threshold, A condition where laser emission just starts to occur; Mie scattering, Mie scattering refers to scattering from spherical nanoparticles that are almost the same size as the wavelength of light; Anderson localization, A consequence of quantum reflection in the lattice that stops the wave function. Localization in this context means the wave would decay and would not be scattered out of the lattice; Scattering mean path,

The average distance a wave travels between collisions; Transport mean path, The average distance a wave travels before its direction of propagation is randomized

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