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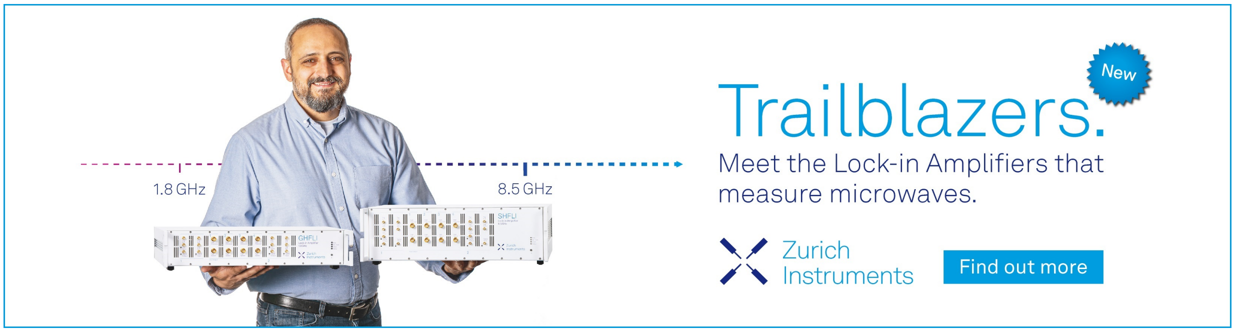

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Principles and Characteristics of Random Lasers and Their Applications in Medical, Bioimaging and Biosensing

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Abstract. A random laser is an optical device that depends on multiple light scattering and gain to provide the feedback mechanism and light amplification respectively. A random laser is different with a regular laser where the regular laser needs a cavity for the feedback mechanism. Multiple light scattering can be considered detrimental in the regular laser but in the random laser, multiple light scattering can increase the path length of light in the active medium. This article studies the operation principles and characteristics of random lasers including the history and applications of the lasers. The theories of light scattering and light amplification are properly explained, followed by a discussion on characteristics of random lasers in terms of random laser emission, emission linewidth and coherence. Furthermore, the unique characteristics of random lasers can contribute to many applications leading to promising element for the future medical and biosensing development.

HISTORY OF RANDOM LASER

In 1968, random laser systems were first theoretically proposed using the diffusion equation by Letokhov *et al.*[1]. The random laser system was then further investigated by the Ambartsunyam *et al.*[2] using a mirror and a surface filled with two ruby crystals as the active gain medium for the feedback mechanism. Then, research on random lasers were continued by Lawandy *et al.* [3] in 1994 through experiments and discussion. They observed a narrowing of the spontaneous emission spectrum when the scatterer's density was over a certain value. The phenomenon was named as a laser-like emission which related to the original work of Letokhov [1]. Lawandy *et al.* [3] shows that random lasers produce high emission intensity, spectrally narrow emission linewidth and consists of the laser-like light emission through optical feedback from multiple scattering.

OPERATING PRINCIPLES OF RANDOM LASERS

Random lasers are unique lasers which depend on multiple light scattering and optical gain. Operation principle of random laser is similar with the regular laser, which the main elements consist of amplification and feedback[4]. The regular laser uses an optical cavity consists of a mirror and a partial reflector to provide the feedback mechanism while light amplification is generated by the pumped gain medium. When gain exceeds loss, the lasing appears through the partial reflector. In random lasers, the feedback is provided by multiple light scattering. Figure 1 depicts the comparison of a regular laser with a random laser.

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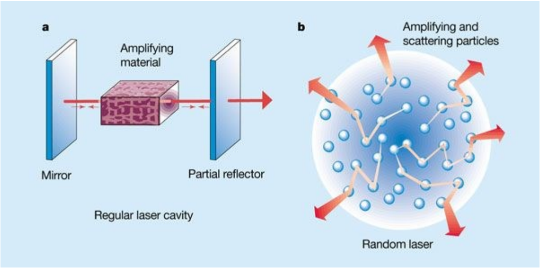

FIGURE 1. a) A regular laser b) A random laser [5]

Multiple Light Scattering

Multiple light scattering is a phenomenon that exists in almost all optical materials that seem hazy such as mists, white paint, powders and even human tissues [6]. The light rays appear to be in multiple scattering whenever they penetrate the opaque materials and being scattered randomly in thousands of time before exiting the materials[6]. Random lasers are known as cluttered optical structures in which light waves are both multiply scattered and amplified [7]. Light waves inside such opaque materials perform random walks, as they are dispersed a few times in irregular ways before leaving the material and resulting the formation of white appearance [7]. These multiple light scattering processes create a perplexing impedance design known as speckle without compromising the coherence of light [7].

There are two types of random lasers, which are incoherent and coherent feedback [8]. Incoherent random lasers work when the light returns to the gain medium by multiple scattering. The photon density rapidly grows with the increase of the pumping rate and a narrowed emission peak is formed on the broad fluorescence background at gain spectrum's center during threshold [8]. Meanwhile, the coherent feedback in random laser occurs when the scattered emission light produces the closed loop paths due to repetitive light scattering which result in emerging of spectral narrow peaks above the emission background when lasing threshold occur [6].

Scattering mean free path, l_s represents an important parameter for the multiple light scattering theory. It refers the average distance that light travels between two successive scattering events. The transport mean free path, represented by l_t, refers to the average distance the light propagates in the sample before the propagation is randomized Theoretically, isotropic and anisotropic scattering exist in random and disordered media. Eq. (1), (2) and (3) are used for isotropic and anisotropic scattering respectively. In isotropic scattering, the transport mean free path is similar to scattering mean free path,l_t~l_s whereas in anisotropic scattering, the transport mean free path is measured based on the scattering angle [9][10].

$$l_s \sim l_t \tag{1}$$

$$l_t = \frac{l_s}{1-(cos\theta)} \tag{2}$$

$$l_s = \frac{1}{\rho\sigma_s} \tag{3}$$

The l_t represents the transport mean free path, while l_s means the scattering mean free path and *cos* θ is the average cosine for the scattering angle[9] [10]. ρ stands for the number density of scatterers and σ_s is the scattering cross section. In the random and disordered media, there are three regimes for multiple scattering using transport mean free path as in eq. (1) [10][11];

(i) Localization regime: $l_t \leq \lambda$
(ii) Diffusive regime: $\lambda < l_t < L$
(iii) Ballistic regime: $l_t \geq L$

The λ represents the light wavelength while L is the sample size in which the lasing takes place.

Scattering can be divided into elastic and inelastic scattering. Elastic scattering occurs when the direction of light propagation is modified, and the energy of the photon is conserved whereas inelastic scattering changes the direction of light and the photon energy. Elastic scattering can be described using Rayleigh, Mie and Geometrics Optics modeling. These three types of modeling depend on the size of nanoparticles. Rayleigh scattering model is used for the elastic light scattering with the diameter of the particles is much smaller than the wavelength of the scattered light[11]. Meanwhile, Mie scattering model is used to analyze the particles for spherical nanoparticles that has the size almost the same to the light wavelength and the Geometrics Optics studied the particles with much larger radius than the light wavelength [11]. The equation (4-6) shows the equation for scattering cross section used for the three types of scattering modeling [11];

Rayleigh, $\boldsymbol{\sigma_{SR}}$;

$$\sigma_{SR} = \frac{8\pi}{3}\left(\frac{2\pi n_m}{\lambda}\right)^4 \alpha^6 \left(\frac{m^2-1}{m^2+2}\right)^2 \tag{4}$$

λ represents the light wavelength, a is the particle radius and $m = n_p / n_m$ is the ratio of the refractive index of the particles to that of the surrounding medium.

Mie, $\boldsymbol{\sigma_{sM}}$;

$$\sigma_{sM} = \left(\frac{2\pi}{k^2 m}\right) \sum_{n=1}^{\infty} (2n+1)\left(|\alpha_n|^2 + |b_n|^2\right) \tag{5}$$

The a_n and b_n are the scattering coefficients which can be found in ref.[11] while $k_m = 2\pi n_m / \lambda$.

Geometrical Optics, $\boldsymbol{\sigma_{sG}}$;

$$\sigma_{sG} = \pi\alpha^2 \tag{6}$$

Where a is the particle radius

Optical Gain

Optical gain is one of the elements needed for the random lasing system besides multiple light scattering. The gain material becomes active when it is excited by a high energy pump source. Light may interact while propagates with an amplifying or gain material. Lasing action takes place when the light is sufficiently amplified before it leaves the framework, providing that gain overcomes loss, [12]. The light waves are multiply scattered and amplified in an amplifying disordered medium. The amplification process is described by the eq. (7) as l_g represents the gain length while l_{amp} refers to the amplification length. The amplification length is defined as the average distance between the beginning and ending point for paths of length, l_g while the gain length refers to the path length over which the intensity is amplified by a factor *e*[13].

$$l_{amp} = \sqrt{\frac{l_t l_g}{3}} \tag{7}$$

As light travels in a straight line in a medium without scattering, the eq. (8) defines that amplification length should equal to the gain length.

$$l_{amp} = l_g \tag{8}$$

Random lasing works the best upon the availability of sufficient scattering and the scattered light is well amplified to balance the loss as it leaves the gain medium[10]. The emitted photons propagate and being amplified within the active gain region, until they escape from the region. Thus, to balance the loss before photons escapes the medium, the equation (9) represents the necessary condition of random lasing so that the light is adequately amplified [9][10].

$$l_s \geq l_g \tag{9}$$

CHARACTERISTICS OF RANDOM LASERS

Random Laser Emission

There are two different spectral signatures of random lasers: incoherent and coherent emission. Coherent/resonant emission is characterized by multiple narrow emission peaks with sub-nanometer linewidth while the incoherent/nonresonant emission deals with a single-peak spectrum of a few nanometers, and both are randomly distributed in frequency[9].

Coherent random laser achieves the lasing when multiple narrow emission peaks appear [13] [14][15]. The random laser and regular laser share similar characteristics in which the emission spectrum can be tremendously narrow, thus producing well defined color and having pulse-like output [8]. When a photon propagates in the gain medium and stimulate the second photon, light will emit from the gain medium. The gain length at frequencies near the maximum of the gain spectrum approaches the average path length of the photons in the gain medium[13]. Therefore, the probability of a photon to generate the second photon will approach one, showing the abrupt increase in the emission intensity. This sudden rise of emission intensity at the maximum of the gain spectrum creates a narrow emission peak [14][16]. Figure 2 shows the narrow emission linewidths of Rh6G / silver random lasers for increasing pump energy density[17].

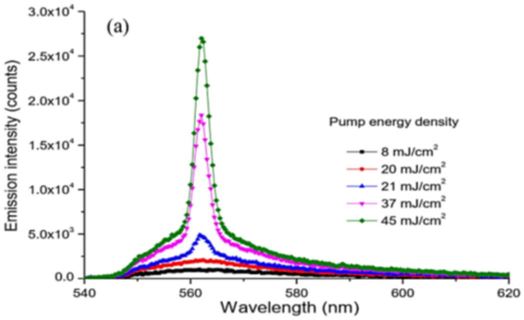

FIGURE 2. The narrow emission linewidths of Rh6G / silver random lasers for increasing pump energy density[17].

The multiple emission peaks with sub-nanometer linewidth can be formed on the top of wide fluorescence background whenever there is increment in scattering [16]. In the weakly scattering regime, multiple emission peaks can be observed from random changes of the refractive index inducing waveguiding and resonant modes with large transport mean free paths[10]. Meanwhile, the spiky emission only can be observed depending on resolution of the detection system, characteristics of the nanostructures and pump pulse duration [6][15][18].

Furthermore, the emission wavelength in random laser can be shifted and tuned [19][20]. The emission wavelength can be tuned by varying the particle/structure size and absorption which changes the gain curve and mode selection [21]. Besides, the emission wavelength also can be tuned by varying other parameters such as temperature, electric field and pump beam distribution [20]. By changing the refractive index of the liquid crystal, the emission wavelength of a random laser with liquid crystal can be tuned by applying a small voltage to the sample to shift the wavelength [22].

Lasing Threshold

Lasing threshold is an important characteristic in random laser. Lasing threshold of the lasers depends on the luminescence efficiency of the gain media and the scattering mean free path of photons in the random media[10]. Lasing threshold is reached when the pump transition is saturated (bleached). The lasing threshold can be determined at the onset of the nonlinear increase of emission intensity as a function of pump energy density [10]. ZnO random laser had been studied to observe the decrease in lasing threshold when the scattering mean free path is equal or lower than the stimulated emission wavelength [10].

In a regular laser, lasing threshold is determined at the beginning of the nonlinear increment of output power as a function of input power [9]. Meanwhile, in a random laser, a lasing threshold happens during the transition of the pump. The lasing threshold can be determined at the onset of the nonlinear increase of emission intensity as a function of pump energy density [9]. There are several elements which can affect the performance of lasing threshold such as the concentrations of the scatteres, the refractive index (RI) of the scatters in comparison with the surrounding media, the excitation spot diameter, energy transfer and the dye concentration[10]. Particles and dye concentration of a material affect the lasing threshold of a random laser [23][24][25].

In 2003, Burin *et al.* performed an experiment to show that the lasing threshold depends on the light transport length, dye concentration and the pump area [15]. The experiment showed that lasing threshold intensity decreases with the light transport length, and the beam diameter saturated at 200μm or 1mm[15]. The lasing threshold also decreases inversely with the increment of dye concentration[15]. The lasing threshold decreases when there is increment in either one or both dye and particle concentration[16].

On the other hand, Cao *et al.*[25] proved that the lasing threshold can be reduced when the scattering mean free path of photons in zinc oxide, is less than the stimulated wavelength. The lasing threshold also can be reduced when there is an increment of the scatterers [24], higher available gain [23] or larger refractive index contrast between the scatterers and the surrounding medium [26]. Figure 3 shows the lasing threshold is reached whenever there is an abrupt change in the transition pattern[17].

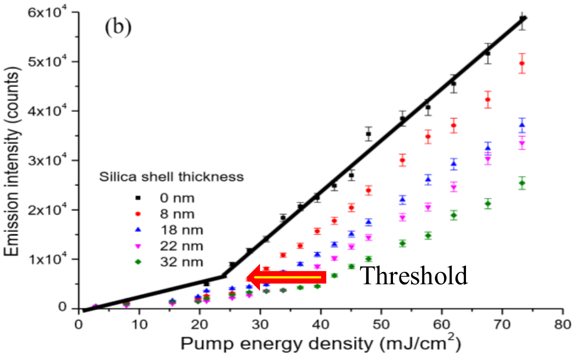

FIGURE 3. The lasing threshold is reached whenever there is an obvious surged in the transition pattern [17]

Coherence

Coherence of the laser radiation is the main key that differentiate between a laser and other light sources [10]. Optical coherence defines the production of interference effects by the ability of light [27][28]. Coherent light exists when there is a fixed phase relationship between the electric field values at disparate locations or times. If there is partial correlation between phase values, the partially coherent light exists [28]. Both random laser and regular laser share the similar spatial and temporal coherence, which spatial coherence is the one that allows the laser beam to focus on a small spot while the temporal coherence causes the laser's spectrum to be very narrow[10].

Coherent source is said to have a Bose-Einstein photon distribution whereas Poisson photon distribution is conceived by the incoherent sources [10]. Cao *et al.* had conducted an experiment to study the changes of photon's distribution from Bose-Einstein to the Poisson distribution in a coherent random laser when the threshold is reached [10][29]. Furthermore, the same experiment repeated for the incoherent Rhodamine 6G-TiO_2 system showing that the random laser can be partially coherent, which combines the Poisson and Bose-Einstein distribution together based on photon statistics [10]. The experiment on disordered nano-structures with both strong and weak scattering to investigate the coherent characteristics of random laser, which then shows that spatial coherence uses the Young's two slit interferometric scheme while the temporal coherence uses a Twyman-Green interferometer and a Michelson interferometer set up [10][30]. W Z W Ismail *et al* [8] also studied coherence of random lasers based on temporal and spatial coherence where the interference fringes and intensity profile above lasing threshold are shown in Fig. 4.

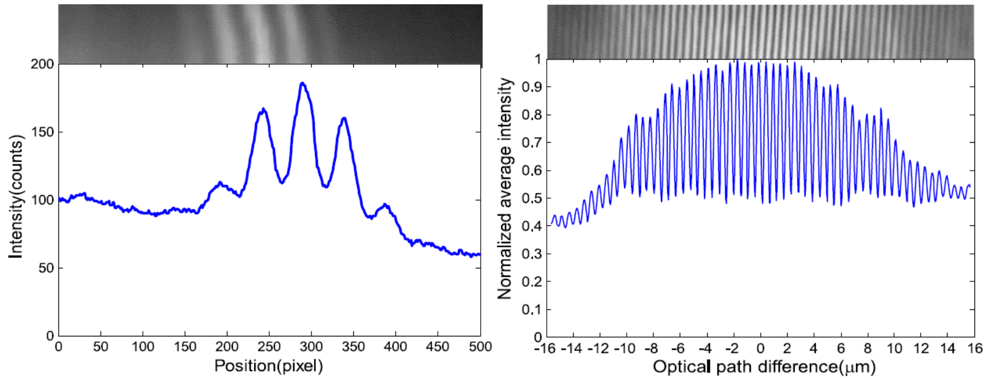

FIGURE 4. The interference fringes and intensity profile for spatial and temporal coherence [8]

APPLICATION OF RANDOM LASERS

Random lasers can be applied in many fields due to their unique characteristics; low fabrication costs, a specific wavelength of operation, flexible shape and substrate compatibility[31][32]. Redding *et al.*[33] has demonstrated that random lasers suitably used for bioimaging as it can produce high-intensity and speckle-free light leading to a creation of a compact on-chip random spectrometer [34].Random lasers also appear to be the best in the image quality test overpowering all the other light sources [34]. Speckle generation tests, image quality tests and contrast-to-noise ratio (CNR) tests had been used to compare the performance of random laser from other light sources. Random laser and LEDs formed the non-speckle patterns in the speckle generation tests and producing higher CNR in the CNR tests. However, random laser appears to be the best in the image quality tests out of all other light sources[33].

Random laser plays an important part in the medical area to detect tumour and do photodynamic therapy[31]. Song *et al.* had reported that random lasers can be used for the sensing purposes[34]. In [34], a 690 nm light can be used to excite the samples of bone which had been soaked with a dye emitting at 800 nm. The characteristics of random lasing in the bone's specimens lead to the detection of nanoscale structural alterations in a mechanical biosensor [34]. On the other hand, W. Z. Wan Ismail *et al.* had demonstrated an impressive application to measure a very low dopamine concentration through aggregation of gold nanoparticles, enhanced by copper ions for random lasers with incoherent feedback which applies emission peak shift, emission peak linewidth, signal-to-noise ratio and lasing threshold with a dopamine detection limit of $\sim 1 \times 10^{-7}$ M as the detection indicators [35].

Besides, the shifts of the emission peak wavelength of random lasers happens when the bone specimens being pressured while undergoes the mechanical testing. Hui Cao mentioned in [32] that Z.V.Vardeny *et al.*[36] has proven the human tissues possess the strong scattering and able to support the random lasing whenever being penetrated with the concentrated laser dye solution. Random lasers had been used to map cancerous tissues whenever the scattering properties of the cancerous tissues portraying the affected spectral emission. The experiments used both healthy and cancerous tissues where the tissues were soaked separately in Rhodamine 6G dye and flattened between two parallel microscope slides. The emission spectra were recorded after the samples were pumped with Nd:YAG laser (532 nm, 100 ps and 800 Hz) and in order to distinguish the tissues, the power Fourier transform (PFT) of the emission spectra was used in the experiments [36].

CONCLUSION

In conclusion, a random laser is an optical device that brings a lot of benefits to human due to its unique characteristics and features. In this review, the main operation elements of random lasers are explained in details where random lasers depend on multiple light scattering and excited gain medium. The characteristics of this laser is also discussed clearly supported by some applicable figures. The uniqueness of random laser which relies upon the coherent and incoherent feedback making this laser suitable for many applications in biosensing, medical and bioimaging. Furthermore, the characteristics of random lasers also can generate unique and sophisticated applications hence becoming the promising element for the development of the future.

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