Spatial coherence measurement of random laser

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Abstract

A new method, double-grating interferometer, is used to investigate the spatial coherence of a random laser based on rhodamine 6G dye and TiO$_2$ nano-particles doped in methanol solution. First, we have observed the lasing behaviour for a series of concentrations of two different diameter nano-particles. We have studied the variation of random lasing with fill factor, which is related to transport free path, and the scatterer diameter. Later, for particular active disordered medium we determined the visibility to observe the spatial coherence of random laser. We have found that random laser source is spatially partially coherent which is in good agreement with other results determined by classical experimental approaches.
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In 1960s, Letokhov first proposed theoretically laser like emission from scattering particles with negative absorption [1, 2]. He introduced this theory following the N. G. Basov et al. experimental work with a new type of cavity having one scattering surface [3]. But, this phenomenon of light amplification was first observed experimentally from powder of laser crystals by Briskina and co-workers in 1986 [4]. They showed that, when the pumping intensity is higher than the threshold, nanoparticles provide a narrow spectrum with peaks. Till early 1990s, there was doubt whether the feedback is brought from total internal reflection or multiple scattering, because the nano-particles serve as both gain and scattering medium [5].

In 1994, a revolution came to the research on random laser when Lawandy described the spectral and temporal properties of radiative emission from laser dye solution with strongly scattering medium [6]. In his experimental approach, the scatterers and the gain medium were independent. The uniqueness of his method is that one can change the concentration of the disordered medium by varying the particle density. This laser like phenomenon in random media is later mentioned as random laser [7, 8]. It was proposed by Florescu et al. that the emission spectra above threshold shows poissonian statistics and this was first experimentally proved by Cao et al [9, 10].

Later on, Jiang et al. developed the time dependent theory for electronic population in 1D random media [11] and Sebah and coworkers propose the rate equation in 2D random media [12]. Enormous amount of theoretical and experimental work had been done at the end of 20th century. At the beginning of twenty first century, Cao classified the random laser into two classes depending on lasing feedback: co-
herent random laser and incoherent random laser [5]. Cao first proposed that the random laser had coherent behaviour and she experimentally showed the coherent properties of random lasing from ZnO\textsubscript{2} powder. Cao proposed that in strong scattering medium with gain, light is confined through recurrent processes and supplies coherent feedback [13]. Andreasen et al. demonstrated theoretically and numerically the characteristics of lasing modes in random system [14]. Noginov et al. and Lui et al. studied the nonlinear effects of random laser. They demonstrated the variation of frequency and structure of lasing mode [15]. Recently, Lopez and coworkers scrutinized the spontaneous mode-locking in disordered resonators which led to the development of a miniature laser [16]. Cao et al. first proposed the possibility of making micro-random laser using disordered strong scattering media. And later they fabricated micro-random laser with ZnO nano-particles [5].

The invention and the development of random laser opened a new era of technological applications. Random distributed feedback fiber laser promises new opportunities in telecommunication and distributed sensing [17]. Micro-random laser is the smallest light source ever that can be used inside an electric circuit or that can be used as optical tags. In medical applications, it can be used to detect cancer, tumours and for photodynamics therapy. It has the potential to be used as a sensor and in machine vision systems [5]. Due to the complex mechanism of random laser, there are many more opportunities to do research in this field. Such as design and engineering of its characteristics, control pumping and lasing mode characteristics.

In this thesis, I determine the spatial coherence of R6G doped TiO\textsubscript{2} random laser. First, I made a series of random media with different concentrations for two size of particles. Then, I investigate the random lasing and its properties. Later, I determine the coherent properties of random lasing.

In chapter two, I will describe the fundamental concept and the theory of random laser, general coherence theory and theory of the grating interferometer. In chapter three, I will talk about the materials and methods. Chapter four follows with the experimental setup and measurement, while chapter five includes results and discussion. Finally, in chapter six I will present the conclusion of my work.
In this chapter I will explain the basic theory of random laser, general concept of coherence and the theory of the double-grating interferometer.

2.1 Theory of Random Laser

We need the basic knowledge of a conventional laser to get better understanding of the random laser. In this section, I will describe the basic properties and the principle of random laser. Also, I will briefly explain how the random medium is formed and how the lasing phenomenon occurs.

A laser is an optical device that emits light that is amplified by stimulated emission. The three basic components of a laser are gain medium, resonator and pump. The resonators confine the light inside the gain medium. The gain medium is pumped with an external source. When the amplification of light overcomes the total cavity losses, lasing occurs [18].

In a random laser the lasing feedback comes from a disordered medium as a result of multiple scattering. Here the scattering particles act to form the cavity instead of a conventional mirror. If there are some scatterers instead of cavity mirrors the light in the gain medium are scattered thousands of times in random directions before leaving the medium. Through this process the scatterers work as a cavity to trap the light in the gain medium. There are two length scales that are related to the random lasing: path length and generation length. The average distance a photon travels before leaving the gain medium is the path length and the average distance it travels before generating a second photon is the generation length. In a disordered medium the path length is increased due to the multiple scattering,
and when it exceeds the generation length, a new photon is generated. This unique process of optical feedback by multiple scattering is called random laser. During the random walk of light within the scattering medium, interference effects appear, which regulate the mode architecture. In a conventional laser, modes are produced by the laser cavity whereas in a random laser modes are determined by multiple scattering [18, 19]. Figure 2.1 shows the schematic diagram of a conventional and a random laser.

There are two types of random lasers (i) coherent random laser, (ii) and incoherent random laser. The coherent random laser is based on field or amplitude feedback but incoherent random laser is based on intensity or energy feedback. In incoherent random laser the scattering mean-free path is greater than emission wavelength. On the other hand, in coherent random laser the magnitude of scattering mean-free path and emission wavelength are of the same order [20, 21].

Few decades ago, there were many myths behind the light scattering from marble stone of Taj Mahal during the moonlight night. But nowadays, its clear that it is due to the disordered structure of stones. The medium where the molecules or ions are randomly oriented and where the propagating light scatters in random direction is called random or disordered medium. It can be constructed either by removing elements from a periodic system or by suspending particles in a liquid medium where the particles can move randomly [19, 22]. As light propagates through a disordered
medium, it experiences multiple scattering, and during this scattering process the scattered light changes its direction in a random way. Therefore, light travels a certain distance before leaving the gain medium [18].

When light ray passes through a disordered medium it can experience three possible events: reflection, transmission and absorption which depend on particle size, density of the medium and intensity of the incident light. In the case of elastic scattering the wavelength of incident light ray remains the same after scattering but it changes during inelastic scattering. Furthermore, single scattering occurs when particle density is low and incident light intensity is high. If the medium density is high or particle size is large, the photon experiences multiple scattering before leaving the medium. In the event of random lasing, the most important multiple scattering phenomenon is back scattering.

When self-generation of photon takes place due to the spontaneous emission of ions or molecules by the excitation of a pump light source, the photon experiences multiple scattering and travels a random distance before escaping the gain medium. There are some length scales that are related to the photon generation. One is generation length and another is path length. Before generating a new photon by stimulated emission, the average distance that is traveled by a photon is called generation length \( l_g \) and the distance that is traveled before leaving the gain medium is called path length \( l_p \) [18]. There are some other path lengths that are related to the random mechanism which are shortly explained below.

The mean free path \( l \) can be expressed as

\[
l = \frac{1}{\beta} = \frac{L}{\ln(I/I_0)}
\]  

(2.1)

where \( I_0 \) is the pump intensity, \( I \) is the transmittance intensity of host medium with gain materials and \( \beta \) is the scattering cross section, which can be obtained by approximating \( \frac{I}{I_0} \) using Lambert-Beer law and \( L \) is the thickness of the sample [23].

To get a better understanding of the operation principle of a random laser, we need to know some physical characteristics of the scattered light ray in the random medium, such as transport mean free path, scattering mean free path, excitation mean free path and diffusive absorption length. These parameters are described below.

The average distance after which the light ray changes its initial direction is
called the transport mean free path and is denoted by $l_t$. It can be written as

$$l_t = \frac{1}{\rho \sigma_t}$$

(2.2)

where $\rho$ is the density of particles and $\sigma_t$ is the transport scattering cross section [24]. Whereas, scattering mean free path is the distance between two consecutive scattering actions, at which the intensity of photon flux scales down by a factor of $1/e$ and is denoted by $l_s$. We can express this as

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

(2.3)

where $\sigma_s$ is the scattering cross section [24, 25].

The gain length ($l_g$) and the amplification length ($l_{amp}$) are used to describe how the stimulated emission causes the light amplification. The light is amplified by a factor of $e$ after a certain distance which is known as the gain length. The amplification length is determined by taking the root mean square of the distance between initial and final points of gain length. If there is no scattering in the medium then light transmits following a horizontal line, thus the gain length and amplification length are equal. So, if the medium is diffusive we can write

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

(2.4)

where $d$ is the diffusion coefficient which can be expressed as $\frac{d t}{v}$ in a 3D system and travelling time, $t = \frac{v l_g}{v}$ [5]. Likewise, if the intensity is reduced by a factor $\frac{1}{e}$ due to absorption, the path length travelled by a photon is then called inelastic mean free path ($l_i$) which is an analogue to the gain length, and it’s written as

$$l_i = \frac{1}{\rho \sigma_a}$$

(2.5)

where $\sigma_a$ is the absorption cross section [24]. Moreover, in the gain medium, the average distance that is travelled by a photon between the initial and the final points of the inelastic mean free path is known as diffusive absorption length ($l_a$) and is given as

$$l_a = \sqrt{\frac{l_t l_i}{3}}$$

(2.6)
Maximum of the photons experience multiple scattering in the medium if $l_a$ is much larger then $l_t$ [24, 26]. Thus, the excitation mean free path can be defined as

$$l^* = \left( \frac{1}{l_i} + \frac{1}{l_a} \right)^{-1}$$

(2.7)

which describe the decrease of peak intensity [24].

The transport mean free path and the scattering mean free path are connected by the relationship

$$l_t = \frac{l_s}{1 - \langle \cos \theta \rangle}$$

(2.8)

where $\langle \cos \theta \rangle$ is the average cosine of the scattering angle. If $\cos \theta=0$ i.e. $l_i=l_s$ for an isotropic medium, photons experience Rayleigh scattering. If $\cos \theta=0.5$ i.e. $l_t=2l_s$ photons experience Mie scattering [27].

In the case of coherent scattering, scattering angle is ignored, because the scattered photon direction is adjacent to the pump beam direction. In the Rayleigh scattering regime, the size parameter, $x = \frac{2\pi r}{\lambda}$, where $r$ is the radius of the scattering particle and $\lambda$ is the pump pulse wavelength. The scattering cross section can be defined as

$$\sigma_{sc} = \frac{8}{3} x^4 \left( \frac{n^2 - 1}{n^2 + 2} \right)^2 \sigma_g,$$

(2.9)

where scattering geometrical cross section $\sigma_g = \pi r^2$. When the fill factor is unchanged but scatterer diameter is reduced, the medium density can be defined as $\rho = \frac{3f}{4\pi r^4}$. Thus, equation (2.9) reduces to [28]

$$l_s \simeq l_t \simeq \frac{\lambda}{4f \pi^4} \left( \frac{\lambda}{d} \right)^3 \left( \frac{n^2 + 2}{n^2 - 1} \right)^2.$$

(2.10)

However, there are three regimes in a random medium. For $l_t \sim \lambda$ the regime is the localization regime. For $L \gg l_t \gg \lambda$ and $l_t \geq L$ the regimes are the diffusive and weakly scattering regime, respectively, where $L$ is the diameter of the system. On the other hand, when $l_a \gg l_t$ and $l_a \ll l_t$ the regimes are strongly and weakly scattering, respectively [27].

2.1.1 Cavity formation in gain medium

Two fundamental elements of a laser are the cavity and the gain medium. The cavity is necessary to trap the light ray in the gain medium for providing positive
feedback. Whereas, the gain medium contains the material that amplifies light by stimulated emission. In a conventional laser, the Fabry-Perot cavity is most familiar, where two mirrors are placed in parallel and light ray travels back and forth between them and becomes amplified. As the light rays propagate, they interfere constructively following the phase delay $2n\pi$. In such a laser, the gain medium is amplified electrically or by using an external laser source. The condition for constructive interference at resonance frequency is

$$kl_{cav} + \varphi_1 + \varphi_2 = 2n\pi \quad (2.11)$$

where $l_{cav}$ is cavity length, $\varphi_1$ and $\varphi_2$ are the phase terms related to the two mirrors and $n$ is an integer.

On the contrary, if some scattering particles are used instead of cavity mirrors, light can interact within the gain medium with the particles and randomize its direction. In such a medium light experiences scattering several times before leaving the medium. Due to multiple scattering some of the light returns to its initial position from where it scattered first, forming a closed loop. Consequently, the dwell time of light increases, i.e. light stays for a longer time within the gain medium which strengthens the amplification. So, scatterers play the same task of trapping light, as cavity mirrors do. Thus, in a random laser, a cavity is formed by disordered materials in the gain medium as a substitute of conventional mirrors [5, 18].

Large scatterers provide a long scattering cross-section which enhances the cavity quality factor for short resonant wavelength and vice versa. With the increasing particle size the transport mean free path and cavity length decreases. Of course, there is a limit above which the transport mean free path becomes constant but emission intensity declines [21]. Nevertheless, for small particle size, the threshold becomes low, but for large particle size its threshold follows a plateau. Also, it is experimentally found that for a certain range of particle size the threshold remains almost the same but a small decrease in lower limit of radius causes a drastic increase in threshold. Cao et al. showed the effect of particle size in transport mean free path ($l_t$). They showed that $l_t$ decreases with particle size and becomes constant for a range of particle diameters, but after a certain level of particle size, $l_t$ increases again. Thus, for a particular range of scatterer size, the transport mean free path exhibits a low value which leads to maximum lasing [28].

In general, light faces a barrier during propagation in a random disordered
medium due to the energy localization. When the wavelength ($\lambda$) of light is smaller than the transport mean free path ($l_t$), light can easily travel through the disordered medium. But, when they are comparable to each other, a localization system is introduced, which gives rise to the interference phenomena. This is the basic concept of Anderson localization, that is analogous to the Ioffe-Regel condition, $\frac{\lambda}{l_t} = 2\pi$ [29]. Moreover, the light localization occurs in a disordered medium under the condition $kl_t \sim 1$; where $K$ is the wave vector magnitude. The coherent backscattering in a disordered medium satisfies the weak localization condition, $kl_t > 1$, which is the precursor of Anderson localization. In a strongly scattering medium the weak localization of light leads to stimulated emission of photons, which points to random lasing [30].

In the case of a typical laser, light rays inside the cavity interfere constructively at some resonance frequency. As a result, light rays remain for a long time in gain medium and get amplified. When the amplification is considerably large to overcome the cavity loss at some frequency, lasing occurs. But in the case of a random laser, the lasing process is quite complex. It is easiest to explain the lasing action of a random laser on the basis of path length. When the scattering medium is dense enough that the path length ($l_p$) becomes large enough to exceed the generation length, ($l_g$) the gain medium reaches a threshold at $l_p = l_g$, and hence commences to generate photons. But, photon explosion appears when $l_p > l_g$ and this type of lasing feedback is incoherent. In other case, when the light comes back to its initial position due to multiple scattering, the scattered light forms a cavity. In such a cavity, due to large amplification, gain exceeds the losses. Therefore, lasing occurs and this type of feedback is coherent.

### 2.1.2 Coherent backscattering

When a light wave is incident on a disordered medium, it experiences multiple scattering. Every single scattering event changes its direction and leaves the gain medium in the backscattering direction by forming a closed loop or in some random direction. This multiple scattered light contains both incoherent and coherent light properties [18]. The scattered light does not always return to its initial location after one round trip. In such a case, the phase relation is ignored and the feedback provided by this type of scattering is incoherent. Conversely, in a strongly disordered medium the incident light follows a random trajectory in the gain medium and return
to its original position from where it was scattered first. This returning process where light forms a closed loop and leaves the medium in the opposite direction of incident light is called backscattering. In a disordered medium the random direction of a photon can be time reversed and light travels in the time reversed direction [25]. In backscattering direction, each trajectory interferes constructively with its time reversed counterpart [18,19,22]. In the closed loop, light can interfere constructively. But, all the wavelengths inside the cavity do not interfere constructively. Only a particular wavelength, at resonant frequency, can interfere inside the cavity i.e. the coherent random lasing is wavelength sensitive.

Coherent backscattering is also called weak photon localization because it has a correlation between photon localization and phase coherent time reversed paths. From the coherent interference of scattered light we can obtain the characteristics of weak localization. The intensity of the scattered light is increased due to the coherent interference and it is nearly double at perfect backward direction, but intensity decreases for the scattering angle above the critical angle [31]. This coherent interference provides information of random lasing. Figure 2.2 illustrates the schematic diagram of coherent backscattering. To know whether the multiple scattering experiences strong or weak localization it is necessary to determine the width of the backscattering cone [32]. The nature of the constructive interference depends on the scattered waves phase difference as

$$\Delta \phi = \frac{2\pi}{\lambda} (x_2 - x_1)$$

(2.12)

where \((x_2 - x_1)\) is the path length difference, \(x_1 = -\hat{i}_1 \cdot (\hat{r}_n - \hat{r}_1)\) and \(x_2 = -\hat{i}_2 \cdot (\hat{r}_n - \hat{r}_1)\) from geometry. Thus, we can write

$$\Delta \phi = \frac{2\pi}{\lambda} (\hat{i}_2 + \hat{i}_1) \cdot (\hat{r}_n - \hat{r}_1).$$

(2.13)

When \(\hat{i}_2 = -\hat{i}_1\), the phase difference between scattered and its time reversed part is zero. As a result, constructive interference occurs. Using the geometry of figure 2.2 we can write Eqn. (2.13) as

$$\Delta \phi = \frac{2\pi}{\lambda} \theta \sqrt{2l_s s}$$

(2.14)

where \(\theta\) is the scattering angle and \(s\) is the total scattering path length. Constructive interference occurs when the phase difference is small and this condition holds for
\[ \frac{\Delta \phi}{2\pi} \ll 1 \] This condition is satisfied for an angle known as the critical angle \( \theta_c \) below which coherent interference occurs [33].

\[ \theta_c \approx \frac{\lambda}{\sqrt{2lt_s}} \] (2.15)

The line shape of coherent intensity peak is determined by

\[ \delta(\theta) = \frac{3}{16\pi} \left[ 1 + 2\frac{x_0}{l_t} + \frac{1}{1 + ql_t^2} \left[ 1 + \frac{1 - \exp(-2qx_0)}{ql_t^2} \right] \right] \] (2.16)

here, \( q = \frac{2\pi \theta}{\lambda} \) and \( x_0 \approx 0.7l_t \). Equation (2.16) is valid only if the polarization of incident field and backscattered field are exactly same [31].

### 2.1.3 Incoherent feedback

If the disordered medium is weakly scattering or strongly scattering but pump pulse area is small, light can easily escape from the active volume. Light may leave the active volume through the front window or travel to unpumped area. But, after
a certain time some of them return to the active volume and are amplified more. Because of random directionality, the scattered light does not return to the initial position from where it was scattered first. As a result, resonant feedback becomes absent, *i.e.* the spectrum is continuous, and in such a random medium feedback is energy or intensity feedback [5]. For such a diffusive random medium Letokhov derived the equation for energy density $\xi(r, t)$ of photon as

$$\frac{\partial \xi(r, t)}{\partial t} = d \nabla^2 \xi(r, t) + \frac{u}{l_g} \xi(r, t) \quad (2.17)$$

where $u$ is the photon velocity inside the medium.

And the solution of the equation (2.17) is given by

$$\xi(r, t) = \sum_m a_m \psi_m(r) e^{-(d b_m^2 - \frac{u}{l_g}) t} \quad (2.18)$$

where $\psi_m$ and $b_m$ are the eigenfunctions and eigenvalues of equation

$$\nabla^2 \psi_m(r) + b_m^2 \psi_m(r) = 0. \quad (2.19)$$

Above the threshold for energy density, equation (2.18) changes to

$$d b_1^2 - \frac{u}{l_g} = 0. \quad (2.20)$$

Here, minimum eigenvalue is $b_1^2$. If we disregard the shape of the medium for a diameter $L$ of the system the minimum eigenvalue can be approximated as $b_1 \approx \frac{1}{L}$. After putting this value in to equation (2.20) we get a critical volume condition as

$$V_{cri} \approx L^3 \approx \left(\frac{l_l l_g}{3}\right)^{\frac{3}{2}}. \quad (2.21)$$

The energy density rises exponentially as a function of time when the medium volume surpasses the critical volume. The system volume is analogous to the characteristic length, *i.e.* when medium volume is larger then the critical volume, the path length exceeds the generation length and hence, photon self generation occurs. If, however, the medium is a solid medium *e.g.* nano-particle disk or rod, equation (2.21) can be written in the form

$$L_{cri} = \pi \sqrt{\frac{l_l l_g}{3}} = \pi l_{amp}. \quad (2.22)$$
Thus, we can write the critical amplification length for the disk having thickness $L$ as $l_{\text{cri}} = \frac{L}{\pi}$. This means that when $l_{\text{amp}} > L_{\text{cri}}$, the system gain overcomes the losses.

The theoretical model of random laser is quite different in the case of coherent feedback from the incoherent feedback. Incoherent random lasers are based on intensity feedback, whereas coherent random lasers are based on field feedback. In incoherent feedback, phase relation is ignored and thus the interference effects are ignored, which is the basic property of coherent feedback. Under these circumstances, a coherent random laser model based on Maxwell’s equations and an incoherent random laser based on diffusion equation can be used [5].

### 2.1.4 One- and two-dimensional random system

In order to develop the time dependent theoretical model of a coherent random laser, Maxwell’s equations are associated with the rate equations of a 4–level atomic system. The obtained equations are solved by using the method of finite difference time domain (FDTD) method in both one dimensional (1D) and two dimensional (2D) systems. Let us consider a 4–level atomic system where electrons jump from energy level 1 to 4 due to pumping. But, electrons decay rapidly to level 3 through nonradiative decay, obtaining a population inversion between levels 3 and 2. Afterwards, the electron drops to level 2 through the radiation of a photon. Finally, the nonradiative transition occurs very quickly from level 2 to 1. Thus the rate equations can be written as

\[
\begin{align*}
\frac{dN_4(r,t)}{dt} &= W_p(t)N_1(r,t) - \frac{N_4(r,t)}{\tau_{43}}, \\
\frac{dN_3(r,t)}{dt} &= \frac{N_4(r,t)}{\tau_{43}} + \frac{E(r,t)}{\hbar\omega_a} \frac{dP(r,t)}{dt} - \frac{N_3(r,t)}{\tau_{32}}, \\
\frac{dN_2(r,t)}{dt} &= \frac{N_3(r,t)}{\tau_{32}} - \frac{E(r,t)}{\hbar\omega_a} \frac{dP(r,t)}{dt} - \frac{N_2(r,t)}{\tau_{21}}, \\
\frac{dN_1(r,t)}{dt} &= \frac{N_2(r,t)}{\tau_{21}} - W_p(t)N_1(r,t).
\end{align*}
\] (2.23)

Here, $N_1$, $N_2$, $N_3$ and $N_4$ are the population densities in energy level 1, 2, 3 and 4, respectively. $\tau_{21}$, $\tau_{32}$ and $\tau_{43}$ are the lifetimes of energy levels 2, 3 and 4, respectively, and $W_p(t)$ is the pumping rate. The polarization density $P(r,t)$ satisfies the following equation

\[
\frac{d^2P(r,t)}{dt^2} + \Delta\omega_a \frac{dP(r,t)}{dt} + \omega_a^2 P(r,t) = \frac{\gamma_r \epsilon_0^2}{\gamma_c m} [N_2(r,t) - N_3(r,t)] E(r,t) \tag{2.24}
\]
where $\omega_a$ is the centre frequency and $\Delta \omega_a$ the line width of atomic transition from level 3 to 2. Additionally, $\gamma_r = \frac{1}{\tau_{32}}$ and $\gamma_c = \frac{e^2 \omega_a^2}{6 \pi \epsilon_0 mc^3}$. Thus, gain is introduced in Maxwell’s equation by the polarization density as

$$\nabla \times \mathbf{E}(r, t) = -\frac{\partial \mathbf{B}(r, t)}{\partial t}, \quad (2.25)$$

$$\nabla \times \mathbf{H}(r, t) = \epsilon(r) \frac{\partial \mathbf{E}(r, t)}{\partial t} + \frac{\partial \mathbf{P}(r, t)}{\partial t}. \quad (2.26)$$

Emission spectrum can be obtained by taking the Fourier transform of $\mathbf{E}(r, t)$ \[5, 34\].

Now we consider the 2D random medium of size $L^2$ where electro-magnetic field consists of two different polarization patterns, namely: the transverse magnetic (TM) field and the transverse electric (TE) field. In the case of transverse magnetic field Maxwell’s equation can be written as \[5, 34, 35\]

$$\mu_0 \frac{\partial H_x}{\partial t} = -\frac{\partial E_z}{\partial y}, \quad (2.27)$$

$$\mu_0 \frac{\partial H_y}{\partial t} = \frac{\partial E_z}{\partial x}, \quad (2.28)$$

$$\epsilon_0 \epsilon_i \frac{\partial E_z}{\partial t} + \frac{\partial P_z}{\partial t} = \frac{\partial H_y}{\partial x} - \frac{\partial H_x}{\partial y}. \quad (2.29)$$

Therefore, the rate equation can be written as

$$\frac{dN_4}{dt} = -\frac{N_4}{\tau_{43}} + W_p N_1, \quad (2.30)$$

$$\frac{dN_3}{dt} = \frac{N_4}{\tau_{43}} - \frac{N_3}{\tau_{32}} + \frac{E_z}{\hbar \omega_a} \frac{dP_z}{dt}, \quad (2.31)$$

$$\frac{dN_2}{dt} = \frac{N_3}{\tau_{32}} - \frac{N_2}{\tau_{21}} + \frac{E_z}{\hbar \omega_a} \frac{dP_z}{dt}, \quad (2.32)$$

$$\frac{dN_1}{dt} = \frac{N_2}{\tau_{21}} - W_p N_1. \quad (2.33)$$

The term $\left(\frac{E_z}{\hbar \omega_a}\right)\frac{dP_z}{dt}$ provides stimulated emission. The polarization density component $P_z$ obeys the equation

$$\frac{d^2 P_z}{dt^2} + \Delta \omega_a \frac{dP_z}{dt} + \omega_a^2 P_z = \frac{\gamma_r}{\gamma_c m} \left[N_2 - N_3 \right] E_z \quad (2.34)$$
On the other hand, for the TE field, Maxwell’s equations are in the form

\[ \frac{\partial H_z}{\partial y} = \epsilon_0 \epsilon_r \frac{\partial E_x}{\partial t} + \frac{\partial P_x}{\partial t}, \]

\[ \frac{\partial H_z}{\partial x} = \epsilon_0 \epsilon_r \frac{\partial E_y}{\partial t} + \frac{\partial P_y}{\partial t}, \]

\[ \frac{\partial E_y}{\partial x} - \frac{\partial E_x}{\partial y} = -\mu_0 \frac{\partial H_z}{\partial t}. \]  

And the rate equations are

\[ \frac{dN_4}{dt} = -\frac{N_4}{\tau_{43}} + W_p N_1, \]

\[ \frac{dN_3}{dt} = \frac{N_4}{\tau_{43}} - \frac{N_3}{\tau_{32}} + \frac{E_x}{\hbar \omega_a} \frac{dP_x}{dt} - \frac{E_y}{\hbar \omega_a} \frac{dP_y}{dt}, \]

\[ \frac{dN_2}{dt} = \frac{N_3}{\tau_{32}} - \frac{N_2}{\tau_{21}} + \frac{E_x}{\hbar \omega_a} \frac{dP_x}{dt} - \frac{E_y}{\hbar \omega_a} \frac{dP_y}{dt}, \]

\[ \frac{dN_1}{dt} = \frac{N_3}{\tau_{21}} - W_p N_1. \]

The polarization density component \( P_x \) and \( P_y \) obey the equations

\[ \frac{d^2 P_x}{dt^2} + \Delta \omega_a \frac{dP_x}{dt} + \omega_a^2 P_x = \frac{\gamma_r e^2}{\gamma_c m} [N_2 - N_3] E_x \]

\[ \frac{d^2 P_y}{dt^2} + \Delta \omega_a \frac{dP_y}{dt} + \omega_a^2 P_y = \frac{\gamma_r e^2}{\gamma_c m} [N_2 - N_3] E_y. \]

### 2.1.5 Three-dimensional random system

Now we consider a three-dimensional (3D) random medium where the scattering particles are placed in a cubic medium with volume \( L^3 \). The rate equations in lasing levels can be represented as

\[ \frac{dN_3(r, t)}{dt} = W_p N_1 - \frac{\sigma I(r)}{\hbar \omega_a} (N_3(r, t) - N_2(r, t)) - \frac{N_3(r, t)}{\tau_{32}} \]

\[ \frac{dN_2(r, t)}{dt} = \frac{\sigma I(r)}{\hbar \omega_a} (N_3(r, t) - N_2(r, t)) + \frac{N_3(r, t)}{\tau_{32}} + \frac{N_2(r, t)}{\tau_{21}}. \]
Here, stimulated emission is obtained by term $\sigma I / h \omega a (N_3 - N_2)$ and the polarization density follows the equation

$$\frac{d^2 P(r, t)}{dt^2} + \Delta \omega a \frac{dP(r, t)}{dt} + \omega^2 a P(r, t) = \frac{\gamma_r e^2}{\gamma_c m} [N_3(r, t) - N_2(r, t)] E(r, t)$$

(2.46)

The population inversion is ignored at $\Delta \nu \gg 1/\tau_{32}$ where $\Delta \nu$ is width of lasing mode. Thus the relation for population inversion fulfills the condition

$$\Delta N = N_3 - N_2 = \frac{N_1 W_p(\tau_{32} - \tau_{21})}{1 + \tau_{32} \sum_{i=1}^{n} \frac{\sigma_i I_i(r)}{h \omega_i}}$$

(2.47)

If the density of the medium is weak then the term $\tau_{32} \sum_{i=1}^{n} \frac{\sigma_i I_i(r)}{h \omega_i}$ in the above equation is ignored [36].

2.1.6 Theory of random laser in diffusive medium

In a diffusive medium the transport mean free path ($l_t$) is much larger than the observed emission wavelength, but much smaller than the diameter of the sample ($L$) i.e. $L \gg l_t \gg \lambda$. In this medium, every single particle acts as a resonator and the total internal reflection from the surface of a particle forms a lasing mode. This is because of the emission spectra which is smaller than the particle size [5]. When light experiences multiple scattering, the scattered light phase becomes randomized, hence the interference effects can be ignored. Let us consider a plane wave which is incident on the z-axis of a slab sample. The ordinary diffusion equation can be reduced to a one dimensional equation under the condition that the z-dimension is much smaller then the x and y dimensions.

$$\frac{\partial W_p(z, t)}{\partial t} = d_p \frac{\partial^2 W_p(z, t)}{\partial z^2} - \frac{d_p}{P_a} W_p(z, t) + P(z, t)$$

(2.48)

$$\frac{\partial W_e(z, t)}{\partial t} = d_e \frac{\partial^2 W_e(z, t)}{\partial z^2} + f v \sigma_{em} N(z, t) W_e(z, t) + \beta \frac{N(z, t)}{\tau_s}$$

(2.49)

$$\frac{\partial N(z, t)}{\partial t} = f v K_a W_p(z, t) - f v \sigma_{em} N(z, t) W_e(z, t) - \frac{N(z, t)}{\tau_s}$$

(2.50)

here, $W_p$ and $W_e$ are the pump and emission light intensity and $N(z, t)$ is the excited state population density. $\beta$ is the ratio of the spontaneous emission rate in lasing
modes and the total spontaneous emission rate. Along the scattering path of the
diffuse light, \( P(z, t) \) is

\[
P(z, t) = \frac{I_0 \sqrt{\ln 2}}{l_s \sqrt{\pi} \Delta} \exp\left(-\frac{z}{l_s}\right) \exp\left(-\left(\frac{t - t_{\text{peak}} - \frac{z}{v}}{\Delta}\right)^2 \ln 2\right)
\]

where, \( I_0 \) is the incident intensity, pump pulse is maximum at \( t_{\text{peak}} \) and its half width
half maximum is \( \Delta \). However, the boundary condition in the x-y plane is \([5, 24]\)

\[
W_p(-l_c^0, t) = W_p(L + l_c^L, t) = W_e(-l_c^0, t) = W_e(L + l_c^L, t) = N(z, 0) = 0
\]

2.2 Coherence theory

In this section I will describe the general coherence theory and coherence theory of
grating interferometer in both space-frequency domain and space-time domain.

Coherence is one of the fundamental properties of light. This property defines
how optical fields at two spatial points and at two time instance (or at two frequencies) are correlated to each other. The spatial coherence signifies the correlation of
the optical fields at two spatial points and at a fixed time (or frequency).

2.2.1 Coherence in space-time domain

Let us consider an electromagnetic field \( U(r, t) \), which is randomly fluctuating at a
position \( r \) and a time \( t \), and satisfies the wave equation in free space. The complex
representation of the field is

\[
U(r, t) = \int_0^\infty U(r, \omega) \exp(-i\omega t) d\omega,
\]

where

\[
U(r, \omega) = \frac{1}{2\pi} \int_{-\infty}^\infty U(r, t) \exp(i\omega t) dt
\]

is a frequency domain electric field at position \( r \) with frequency \( \omega \). In space-time
domain the mutual coherence function (MCF), denoted by \( \Gamma(r_1, r_2, \tau) \), is used to characterize
the field properties at a pair of positions \( r_1 \) and \( r_2 \), and at a time difference \( \tau = t_2 - t_1 \) as

\[
\Gamma(r_1, r_2, \tau) = \langle U^*(r_1, t) U(r_2, t + \tau) \rangle.
\]
Here the angular brackets denote either time average or ensemble average of the field as
\[
\langle U^*(r_1, t)U(r_2, t + \tau) \rangle = \lim_{T \to \infty} \frac{1}{2T} \int_{-T}^{T} U^*(r_1, t)U(r_2, t + \tau)dt. \tag{2.56}
\]

Also, it is more convenient to represent the ensemble average for a set of field representations \(U(r, t)\) as
\[
\langle U^*(r_1, t)U(r_2, t + \tau) \rangle = \lim_{N \to \infty} \frac{1}{N} \sum_{n=1}^{N} U^*_n(r_1, t)U_n(r_2, t + \tau). \tag{2.57}
\]

Thus, the average intensity, \(\langle I(r) \rangle\), of the field at the point \(r\) can be written as
\[
\langle I(r) \rangle = \langle |U(r, t)|^2 \rangle = \Gamma(r, r, 0). \tag{2.58}
\]

However, it is advantageous to normalize the mutual coherence function to study the space-time correlation of the field, which is known as the complex degree of coherence and is defined as
\[
\gamma(r_1, r_2, \tau) = \frac{\Gamma(r_1, r_2, \tau)}{\sqrt{\Gamma(r_1, r_1, 0)\Gamma(r_2, r_2, 0)}} = \frac{\Gamma(r_1, r_2, \tau)}{\sqrt{I(r_1)I(r_2)}}, \tag{2.59}
\]
where \(\Gamma(r_i, r_i, o)\) denotes the intensity at point \(r_i\) \((i = 1, 2)\). Thus, the fringe visibility of Young’s interference experiment corresponds to the modulus of \(\gamma(r_1, r_2, \tau)\). This satisfy the inequality
\[
0 \leq |\gamma(r_1, r_2, \tau)| \leq 1, \tag{2.60}
\]
which means that the field is completely spatially coherent at two points \((r_1, r_2)\) for the maximum value and is completely spatially incoherent for the minimum value. It can be said that the field is partially coherent when the value is between these two extremes [37].

### 2.2.2 Coherence in space-frequency domain

In the space-frequency domain there is a correlation between spatial coherence and the cross-spectral density function (CSDF). More specifically, the spatial coherence at a frequency \(\omega\) can be determined by the tensor form of cross-spectral density function, \(W(r_1, r_2, \omega)\), which is the Fourier transform of mutual coherence function, \(\Gamma(r_1, r_2, \tau)\), i.e. [38],
\[
W(r_1, r_2, \omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \Gamma(r_1, r_2, \tau) \exp(i\omega\tau)d\tau \tag{2.61}
\]
In a special case, the CSDF can serve as the spectral density, \( S(r, \omega) \), of the field when the two points coincide. Hence we can define spectral density as

\[
S(r, \omega) = W(r, r, \omega) = \langle |U(r, \omega)|^2 \rangle. \tag{2.62}
\]

In the frequency domain, one may introduce the normalized cross-spectral density function as

\[
\mu(r_1, r_2, \omega) = \frac{W(r_1, r_2, \omega)}{\sqrt{S(r_1, \omega)S(r_2, \omega)}}, \tag{2.63}
\]

which is known as spectral degree of coherence and satisfies the inequality

\[
0 \leq |\mu(r_1, r_2, \omega)| \leq 1. \tag{2.64}
\]

The upper and lower limits denote that the field is completely coherent or incoherent at points \( r_1, r_2 \) and at a frequency \( \omega \). Between these limit the field is spectrally partially coherent [38–40].

The complex degree of coherence in space time and space frequency domains are connected by the following relation as

\[
\gamma(r_1, r_2, \tau) = \int_0^\infty \sqrt{s(r_1, \omega)s(r_2, \omega)} \mu(r_1, r_2, \omega) \exp(-i\omega \tau) d\omega, \tag{2.65}
\]

where

\[
s_j(r, \omega) = \frac{S_j(r, \omega)}{\int S_j(r, \omega) d\omega}, \quad (j = 1, 2) \tag{2.66}
\]

And inversely,

\[
\mu(r_1, r_2, \omega) = \frac{1}{2\pi} \frac{1}{\sqrt{s(r_1, \omega)s(r_2, \omega)}} \int_{-\infty}^{\infty} \gamma(r_1, r_2, \tau) \exp(i\omega \tau) d\tau. \tag{2.67}
\]

Thus, Eqn. (2.65) and Eqn. (2.67) show the connection between the MCF and the CSD [41].

2.2.3 Theory of the grating interferometer

Young’s double slit experiment is the most prominent experimental method to measure spatial coherence of a light source. But its main drawback is to measure the coherence of weak fields are challenging. Since, the pinhole diameters are small, the distance between the pinholes and the observation plane from the pinholes are considerably large. Thus, considering all these limitations Professor Jari Turunen
and his PhD student Matias Koivurova introduce a new method, namely the grating interferometer, to determine spatial coherence.

Let us consider the grating geometry of figure 2.3. Where, spatially coherent beam is split into two orders $m = \pm 1$. The beam displacement $\pm \Delta x$ from optical axis is

$$\Delta x = \Delta z \tan \theta = \Delta z \tan[\arcsin\left(\frac{2\pi c}{\omega d}\right)] \approx \pm \frac{2\pi c \Delta z}{\omega d}$$

(2.68)

where $\Delta z$ is the propagation distance and $\theta_{\pm}$ is the propagation direction.

### 2.2.4 Space-frequency domain

We can write the total field at a distance $\Delta z$, by neglecting the diffractive spreading of the orders, as

$$E(x, \Delta z; \omega) = E_0(x - \Delta x; \omega)[T]\exp(i\phi_{+1})\exp[i(k_x x + k_z \Delta z)]$$

$$+ E_0(x + \Delta x; \omega)[T]\exp(i\phi_{-1})\exp[i(-k_x x + k_z \Delta z)]$$

(2.69)

where $k_x = 2\pi/d$ and $k_z = \sqrt{(\omega/c)^2 - k_x^2}$.

The spatial interference pattern can be written as

$$S(x, \Delta z; \omega)/|T|^2 = S_0(x - \Delta x; \omega) + S_0(x + \Delta x; \omega)$$

$$+ W_0(x - \Delta x, x + \Delta x; \omega)\exp(-i4\pi x/d)$$

$$+ W_0^*(x - \Delta x, x + \Delta x; \omega)\exp(i4\pi x/d).$$

(2.70)
From the definition of complex degree of spectral coherence is
\[
\mu_0(x_1, x_2; \omega) = \frac{W_0(x_1, x_2; \omega)}{\sqrt{S_0(x_1; \omega)S_0(x_2; \omega)}}. \tag{2.71}
\]
Thus,
\[
S(x, \Delta z; \omega)/|T|^2 = S_0(x - \Delta x; \omega) + S_0(x + \Delta x; \omega) + 2\sqrt{S_0(x - \Delta x; \omega)S_0(x + \Delta x; \omega)}|\mu_0(x - \Delta x, x + \Delta x; \omega)|
\times \cos\{\arg[\mu_0(x - \Delta x, x + \Delta x; \omega)] - 4\pi x/d\}. \tag{2.72}
\]
Finally, the fringe visibility can be written as
\[
V(x, \Delta z; \omega) = 2\sqrt{S_0(x - \Delta x; \omega)S_0(x + \Delta x; \omega)} \times |\mu_0(x - \Delta x, x + \Delta x; \omega)|. \tag{2.73}
\]
From the position of interference fringes the phase of this quantities can be obtained [42].

2.2.5 Space-time domain

We know the Wiener-Khintchine theorem as
\[
\Gamma(x_1, x_2; \Delta t) = \int_0^\infty W_0(x_1, x_2; \omega) \exp(-i\omega \Delta t) d\omega. \tag{2.74}
\]
At \(\Delta t = 0\), we can write intensity distribution formula from equation (2.70) as
\[
I(x, \Delta z)/|T|^2 = \int_0^\infty S_0(x_1 - \Delta x; \omega) d\omega + \int_0^\infty S_0(x_1 + \Delta x; \omega) d\omega
\times \exp(-i4\pi x/d)W_0(x - \Delta x, x + \Delta x; \omega) d\omega
\times \exp(i4\pi x/d)W_0^*(x - \Delta x, x + \Delta x; \omega) d\omega. \tag{2.75}
\]
The complex degree of spatial coherence of the incident field is
\[
\gamma_0(x_1, x_2; \Delta t) = \frac{\Gamma_0(x_1, x_2; \Delta t)}{\sqrt{I_0(x_1)I_0(x_2)}} \tag{2.76}
\]
where \(I_0(x) = \Gamma_0(x, x, 0)\).
If we ignore the \(\Delta x\) dependence on \(\omega\), we may write equation (2.75) as
\[
I(x, \Delta z)/|T|^2 = I_0(x - \Delta x) + I_0(x + \Delta x)
\times \Gamma_0(x - \Delta x, x + \Delta x; 0) \exp(-i4\pi x/d)
\times \Gamma_0^*(x - \Delta x, x + \Delta x; 0) \exp(i4\pi x/d). \tag{2.77}
\]
In terms of complex degree of coherence we can write equation (2.77) as follows

\[
\frac{I(x, \Delta z)}{|T|^2} = I_0(x - \Delta x) + I_0(x + \Delta x) \\
+ 2 \sqrt{I_0(x - \Delta x)I_0(x + \Delta x)} |\gamma_0(x - \Delta x, x + \Delta x; 0)| \\
\times \cos\{\arg[\gamma_0(x - \Delta x, x + \Delta x; 0)] - 4\pi x/d\},
\]

(2.78)

where

\[
\gamma_0(x - \Delta x, x + \Delta x; 0) = \int_0^\infty \sqrt{S_0(x - \Delta x; \omega)S_0(x + \Delta x; \omega)} \\
\times \mu_0(x - \Delta x, x + \Delta x; \omega). 
\]

(2.79)

Thus, in space-time domain the visibility is related to the absolute value of complex degree of coherence as

\[
V(x, \Delta z) = \frac{2\sqrt{I_0(x - \Delta x)I_0(x + \Delta x)}}{I_0(x - \Delta x) + I_0(x + \Delta x)} \times |\gamma_0(x - \Delta x, x + \Delta x)|
\]

(2.80)

For quasimonochromatic light this conditions are valid [42].

2.2.6 Visibility

It is difficult to get complete information on coherence from the intensity at the measurement plane. Michelson proposed a theory to determine the coherence by measuring the sharpness of fringes, which is known as visibility, and defined as

\[
V(r) = \frac{\langle I \rangle_{\text{max}} - \langle I \rangle_{\text{min}}}{\langle I \rangle_{\text{max}} + \langle I \rangle_{\text{min}}}, 0 \leq V \leq 1
\]

(2.81)

where \( I_{\text{max}} \) and \( I_{\text{min}} \) are the maximum and minimum intensities of interferences fringes. The average intensity \( I(r) \) at a point is given by

\[
I(r) = \langle |V(r;t)|^2 \rangle = \Gamma(r, r; 0).
\]

(2.82)

Assuming that the intensity of the wavefront from the two pinholes are equal, i.e. \( I^{(1)}(r) = I^{(2)}(r) = I^{(0)}(r) \), thus

\[
I(r) = 2I^{(0)}(r)\{1 + \frac{\Gamma(r_1, r_1; \tau)}{\sqrt{I(r_1)} \sqrt{I(r_2)}} \left| \cos[\alpha(r_1, r_1; \tau)] \right| \}
\]

(2.83)
where $I^{(1)}(\mathbf{r})$, $I^{(2)}(\mathbf{r})$ and $I^{(0)}(\mathbf{r})$ are the intensities from $+1$, $-1$ orders and their equivalent intensity respectively. From equation (2.81) and equation (2.83) we can write the visibility of fringes for a spatially homogenous field at the observation plane as

$$V(\mathbf{r}) = \left| \frac{\Gamma(\mathbf{r}_1, \mathbf{r}_1; \tau)}{\sqrt{I(\mathbf{r}_1)} \sqrt{I(\mathbf{r}_2)}} \right|, 0 \leq V \leq 1. \quad (2.84)$$

The visibility of equation (2.84) is related to the complex degree of coherence of equation (2.76). If $V$ is 0, light is completely incoherent, when $V = 1$ light is completely coherent and otherwise it is partially coherent [43].
In this chapter I will discuss the properties and the characteristics of the materials that are used in our experiment on random laser. I will also explain briefly the methods that are used to measure the coherence properties of light.

3.1 Materials

To architect a random medium, various chemical elements are essential where only the gain medium is optically active as lasing material. In the case of a random laser, the sample consists of three different components: (i) host medium, (ii) gain medium and (iii) scattering center. Here, all these components and their functions will be described and I will discuss about other elements that were used in the experiment.

It is very challenging to deal with liquid sample as a lasing medium due to the fluctuation of scatterer concentration. For this reason, organic solvents are used as a host medium in which scattering particles and gain medium (laser dye) are dissolved [5]. There is a long list of organic solvents that are used as a host medium e.g. methanol, ethanol, ethylene-glycol. In this experiment of random laser we used methanol \((CH_3OH)\) as a host medium. Obviously, methanol is an ideal solvent because of its ability to dissolve polar and non-polar components equally.

In random laser, organic molecules are used as a laser dye that acts as a gain medium. There are many molecules that have the ability to serve as a gain medium, for example Rhodamine 6G (R6G), Rhodamine B, Fluorescein 27, Pyrromethene 597, Sulforhodamine B. Among them, Rhodamine 6G is a widely used dye and also used it in this experiment. Of course, there are some important reasons to choose R6G, for example, it produces maximum fluorescence and short peak wavelength
in methanol solution. Moreover, in methanol solution it has highest lifetime. In addition, R6G has more than fifty percent energy conversion efficiency and lasing range between 555 nm to 585 nm [44]. The absorption and the emission cross section of Rhodamine 6G is relatively large but the excited state lifetime is short. More importantly, Rhodamine 6G forms a four level system and its ground level is unpopulated.

To achieve strong scattering in a random medium, it is essential to select the scatterers that can change the refractive index of disordered medium randomly. Low absorption coefficient is analogous to short path length, because low absorption is inversely related to the longest path length. There are a wide range of scattering particles used in random laser experiments, which have high refractive index. The most widely used materials are ZnO, TiO$_2$, SiO$_2$ and Al$_2$O$_3$. In this work, TiO$_2$ was chosen due to its high refractive index ($n \approx 2.8$) which is even higher than ZnO ($n = 2$) and low absorption coefficient which is about 10 mm$^{-1}$ [45]. Also, the emitted photons are not absorbed in the random media by TiO$_2$. Moreover, at room temperature, undoped TiO$_2$ acts as an insulator with high energy band gap $E_g \approx 3$ eV. It is an interesting component because it can show both $n-$ and $p-$ type transition at temperature below 927°C but above this temperature TiO$_2$ behaves as an $n-$type semiconductor. TiO$_2$ can absorb light only below the wavelength of 400 nm due to its wide band gap. The absorption causes the transfer of electrons from full valence band to empty conduction band. Emission of photons occurs as the electrons return to valance band. But, when TiO$_2$ is mixed with other components such as a laser dye, it becomes sensitive at high visible wavelength [46]. In our experiment we used TiO$_2$ nanoparticles having a diameter of 100 nm and 300 nm.

### 3.2 Random lasing technique

There are at least two different possible approaches to produce random lasing. One is solid random media where scatterers and gain medium are powders, which are compressed to form a slab geometry. Another is liquid medium, where scatterers and laser dye are dissolved in a liquid host. In our experiment we used a liquid medium to make a random laser. Here, I will briefly describe the basic principle of both methods.

In order to produce random lasing, the nano or micro particles are composed in a
specific shape which can be a slab geometry or a nano-rod, where scatterers perform as both gain medium and scattering center. This random media can be made by mixing scatterers and laser dye or only by scatterers. In the slab of nano-particle powder, every single scatterer acts as a potential resonator, and due to the total internal reflection from a scatterer surface the lasing mode is formed. From these modes, incoherent lasing occurs, but if the scattering is strong, recurrence appears, which causes coherent lasing. When the pump beam is focused onto the surface of the slab, electrons jump to the conduction band by absorbing photons and afterwards return to the valence band by emitting a photon [5]. When the pumping intensity is low, the emission spectrum shows a wide spontaneous emission peak. But, due to increased intensity, at a certain level it overcomes threshold, hence exhibiting a sharp peak on the top of the spectrum. Further increasing intensity there appear other supplementary narrow peaks [18].

The most popular and widely used random lasing medium is liquid medium, where a host liquid contains nano-particles and a laser dye. In the liquid sample, nano-particles serve as scatterers to form the cavity, whereas the laser dye serves as a gain medium. The advantage of this method is that one can change the scatterer concentration in solution by increasing the density of particles which build the strong scattering medium. In this case, the feedback is scattering dependent, i.e. strong scattering leads to short path length. When a pump pulse is focused on the sample, a dish shaped spot appears and under this area gain medium is amplified. The emitted photons leave the gain medium through the front window or travel to the neighbouring unpumped area. Some photons come back to the pump region again after a long random walk, which provides energy i.e. incoherent feedback. Moreover, it is experimentally proven that threshold decreases more than two orders of magnitude by increasing the scatterer density with three orders of magnitude, at a fixed density of laser dye [5]. This implies that more dense sample provides high amount of multiple scattering events, and hence, large feedback. In fact, the cavity formed by multiple scattering in the disordered medium changes its configuration due to the mobility of particles. Because of this, the frequency of each lasing mode is distinct [5,18].
3.3 Coherence measurement method

Young’s double slit experiment is a classical and well known method to determine the spatial coherence of any light source. This method is also used to measure the spatial coherence of random laser despite of its some limitations. Considering these limitations, a new experimental method introduced by professor Jari Turunen and his PhD student Matias Koivurova, named: Double Grating Interferometer (DGI).

The principle of the double grating interferometer is very simple. Instead of a slit, a grating is used to split the light. In this experimental method two binary phase gratings are placed in parallel and a movable array detector is placed in the $z$-direction. The first grating ($g_1$) period is $d$ and the second grating ($g_2$) period is $\frac{d}{2}$. Back scattered light from a random source is incident on the first grating and diffracted into $0^{th}$ and $\pm 1$ orders. Again, these diffracted light are incident into second grating where $0^{th}$ order is blocked. Thus, $\pm 1$ orders are diffracted again into different orders. Consequently, after the second diffraction, one $+1$ order and one $-1$ order part of light interfere. The moving detector detects the light at different positions, which interfere during the traveling. Figure 2.3 shows the schematic diagram of a double grating interferometer [42].
In this chapter I will discuss about the sample preparation, experimental process of random lasing and coherence measurement.

4.1 Sample preparation

R6G has a molar mass of 497.02 g/mol, solubility in methanol is 400 g/l and its chemical formula is $C_{28}H_{31}N_2O_3Cl$. In my experiment, I suspended required amount of Rhodamine 6G in pure methanol solution to prepare the concentration of 5 mM/l. To get the desired concentration, I dissolved 1 g of R6G dye in 417.5 ml of pure methanol. Later, I suspended various amounts of two different diameters, 100 nm and 300 nm, $TiO_2$ scatterers in the solution to make a disordered medium with different concentrations. The concentration of the sample is prepared by considering some parameters: particle volume, particle density and fill factor. The volume of the scatterers for nano-particle having a diameter of 100 nm and 300 nm are $5.23 \times 10^{-16}$ cm$^3$ and $1.141 \times 10^{-14}$ cm$^3$, respectively. Moreover, I intended to make samples of 8 ml dye solution having the concentration between $10^9$ cm$^{-3}$ to $10^{13}$ cm$^{-3}$. I suspended 4.77 mg, 47.7 mg, 0.48 g and 4.8 g $TiO_2$ nano-particle, having diameter of 300 nm, in 8 ml dye solution to prepare the sample concentration $10^9$ cm$^{-3}$ to $10^{12}$ cm$^{-3}$, respectively. Similarly, to make the concentration between $10^{10}$ cm$^{-3}$ to $10^{13}$ cm$^{-3}$ for 100 nm diameter I suspended scatterers $1.77 \times 10^{-2}$ mg, $17.7 \times 10^{-2}$ mg, 1.77 mg, 17.7 mg and 1.8 g in 8 ml dye solution, respectively. During the measurement I tried to keep maximum accuracy and used sartorius 2432 balance, which has an accuracy of 99.958%. It is to be mentioned that for smaller sized particles, our first sample concentration was $10^{10}$ cm$^{-3}$, because it was not
only challenging, but impossible with our equipment to weigh an accurate amount of particles for a concentration of $10^9$ cm$^{-3}$. Also, for large size particle our last concentration was $10^{12}$ cm$^{-3}$ and I did not prepare concentration $10^{13}$ cm$^{-3}$ due to high fill factor. Furthermore, the fill factor for small size particle are $6.54 \times 10^{-7}$, $6.54 \times 10^{-6}$, $6.54 \times 10^{-5}$ and $6.54 \times 10^{-4}$ between concentration $10^{10}$ cm$^{-3}$ to $10^{13}$ cm$^{-3}$, respectively. Whereas, in the case of large size particles, fill factors are $1.76 \times 10^{-6}$, $1.76 \times 10^{-5}$, $1.76 \times 10^{-4}$ and $1.76 \times 10^{-3}$ for concentration $10^9$ cm$^{-3}$ to $10^{12}$ cm$^{-3}$, respectively.

4.2 Experimental approach of random laser

In this section I will discuss about the experimental setup of random laser and the measurement of random lasing.

The outline of the experimental setup is illustrated in figure 4.1. A frequency doubled Q-switched Nd: YAG laser is placed on optical table having a wavelength $\lambda = 532$ nm. A mirror is placed to reflect incident light toward the sample. The sample cuvette is on top of a small magnetic stirring table and a convex lens is placed at a distance of 50 mm, which is the focal length of the lens. Next, a beam splitter is arranged between the mirror and the lens which splits the backscattered light toward the grating. A filter is used after the beam splitter in order to block the wavelength 532 nm. Likewise, an attenuator and a pinhole is fixed between the reflecting grating and filter so that backscattered beam can be attenuated before incident on grating. Later, another convex lens is placed at 25.4 mm away to focus the reflected light on a sensor. Finally, a Thorlabs DCC 1545M monochromatic camera is connected to a computer to record the spectra.

The pump source repetition rate is 10-Hz and the pulse length is 4 – 6-ns. Also, the pump pulse intensity is $\sim 800$ W/m$^2$ and energy is $\sim 100$ mJ. After switching on the pump laser light is emitted which is directed toward the sample by a mirror. Next, we put a magnetic stirring rod into the cuvette to prevent the sedimentation of scattering center and placed the cuvette on the magnetic table. Before placing the cuvette on table we cleaned its surface so that dust can not make any barrier for back scattered light emission and then we placed it carefully exactly at the focal point of the lens. First, we chose the sample having the concentration $10^{10}$ cm$^{-3}$ with particle size of 100 nm. When light hits the surface of the random medium, there is
Figure 4.1: Schematic diagram of experimental setup for random lasing.

A disk shaped spot on the cuvette surface, which is the width of the pulse. Inside the random media this area becomes pumped, and light experiences multiple scattering. Therefore, back scattered light is emitted in the opposite direction of incident light. After passing through the lens, the back scattered light becomes collimated and its split by the beam splitter towards the filter. Filter blocks the incident pump wavelength (532 nm) and permits other wavelengths to pass. Next, the light from the sample is attenuated and light width is controlled with a pinhole before incident on the grating. The grating period of the reflecting grating is $d = 416.5$ nm and the angle between the incident light and the grating is $\theta_i = 30^\circ$. The diffracted light is focused on the camera by a convex lens and the spectrum is recorded by a computer. In our measurement, instead of images we recorded spectrum videos of duration 15 to 20 seconds, which were later converted to images with a matlab code and with the help of matlab we found the spectrum of random lasing. During the measurement we carefully adjusted the CCD camera exactly to the focal point of the lens. The distance between the grating and the camera is 5.5 cm. Likewise,
following this process, I recorded data for concentration $10^{11} \text{ cm}^{-3}$ to $10^{13} \text{ cm}^{-3}$, respectively, for small size scattering centre. Similarly, in the next step we measured data for different concentrations, $10^9 \text{ cm}^{-3}$ to $10^{13} \text{ cm}^{-3}$, of large scattering centre.

To analyze the data I used a Matlab code (see appendix A) that takes single shot image from where spectra is studied. First, we select the height of the frame from where it will take a cross section. Next, a video reader reads the frames from the file at a particular height. Loop through the video to read each frame into width-by-height-by-3 array data matrix, which are written out in JPG form. Later, convert the set of image names to a cell array. Another loop through the image sequence, load each image, and then write it to a matrix. Finally, an image is formed from where we can find out most promising spectra.

From the image we find the bright spot and at this spot signifies the most promising

![Figure 4.2: Images show the temporal evolution of the spectrum. It also gives information that with time variation the number of cavity changes and consequently the coherence of random lasing.](image)

spectra. Figure 4.2 shows the spectral images of two samples, where there are some bright spots. Thus, these figures show the peaks on the top of ASE spectra from which we can identify is it lasing or not. Figure 4.3 represents the emission spectra of two different concentrations of two different size nano-particles.

The spectrometer resolution was between $2.86 \times 10^{-2}$ to $1.95 \times 10^{-2} \text{ nm}$ for the wavelength range 495 to 570 nm. The detail calculation and discussion are provided in result and discussion chapter.
4.3 Coherence measurement of random laser

In this section I will discuss the experimental setup and measurement approach of spatial coherence of random laser. First, I explain briefly the experimental setup and next discuss step by step how to measure coherence of random laser.

Figure 4.4 shows the experimental illustration of coherence measurement. This setup is the rearrangement of previous experimental setup. Here, we kept the setup same up to sample position, then we rearranged some optical equipments for coherence measurement. Two transmittance gratings were used in this setup. A block
Figure 4.4: Schematic illustration of experimental setup for coherence measurement of random laser.

is attached on the second grating to block the zeroth of the diffracted light. In addition, a DCC 1545 monochromatic camera is placed on the optical table which is on the same line direction of twin-grating. Finally, the camera position is adjusted with a computer to record data.

First, we selected the sample having a concentration of $10^9 \text{ cm}^{-3}$ for large scatterers with a diameter of 300 nm to measure coherence. When the sample was pumped, the coherent light traveled in the backscattered direction and was focused on the first grating by a lens. Also, an aperture was used to control the brightness and size of the focused light beam. The first grating divided the light intensity with equal energy between $+1^{st}$ and $-1^{st}$ orders. The $0^{th}$ order is blocked before incident on the second grating. Later, the $\pm 1$ orders are incident on the second grating and diffracted again into $\pm 1$ orders. One $+1^{st}$ order and one $-1^{st}$ order interfere with each other and travel towards the camera. The camera recorded data at 100 positions and for better accuracy the images were taken three times at every position. After taking one set of data the sample was replaced with another sample having
concentration of $10^{10}$ cm$^{-3}$ for large nano-particle. It should be mentioned that this was done only for these two concentrations for 300 nm particles. Because it was clear from earlier measurements that concentrations $10^{11}$ cm$^{-3}$ and $10^{12}$ cm$^{-3}$ did not lose enough due to high fill factor. Next, the same experimental process was repeated for small size scatterers having diameter 100 nm and concentration $10^{10}$ cm$^{-3}$, $10^{11}$ cm$^{-3}$, $10^{12}$ cm$^{-3}$ and $10^{13}$ cm$^{-3}$, respectively. Finally, the interference fringe images were recorded for empty sample that did not contain any scatterers and only the gain medium was present. The empty sample fringe pattern was determined to compare and examine the variation of random lasing visibility and also to exclude the possibilities of artifact. Figure 4.5 shows the interference fringe pattern for large scatterers, small scatterers and empty sample.

![Image](image1.jpg)

**Figure 4.5:** Interference fringe images of (a) large scatterers (b) small scatterers (c) empty sample.

After taking the images of interference pattern, it is essential to analyze the visibility to determine if the source is coherent or not. To diagnose the visibility a matlab code was used (see appendix B). First, three different height were selected to take cross sections. Thus, there were three sets of images at every position namely: imageA, imageB, imageC, matlab code measured the maximum and minimum intensity at every cross section. Next, averaged over it for three different cross sections and later on the three different sets of images. After getting the maximum and minimum intensity matlab code follow the equation (2.81) to find out the visibility. This way, the matlab code calculated the visibility at every position and also found out the maximum visibility for every concentration. Similarly, I found out the max-
imum visibility of concentration $10^{10}$ cm$^{-3}$, $10^{11}$ cm$^{-3}$, $10^{12}$ cm$^{-3}$ and $10^{13}$ cm$^{-3}$ for small size nano-particle and $10^{9}$ cm$^{-3}$ and $10^{10}$ cm$^{-3}$ for large size nano-particle. Figure 4.6 shows the visibility graph, position vs maximum visibility, of six different concentrations.
Figure 4.6: Maximum visibility vs sample position graph for concentration $10^9$ cm$^{-3}$ (300 nm), $10^{10}$ cm$^{-3}$ (300 nm), $10^{10}$ cm$^{-3}$ (100 nm), $10^{11}$ cm$^{-3}$ (100 nm), $10^{12}$ cm$^{-3}$ (100 nm) and $10^{13}$ cm$^{-3}$ (100 nm) from left top corner to right bottom corner respectively.
In this chapter I will discuss about the lasing process of random laser and its properties, and the characteristics of random lasing. Also, I will discuss about the coherence properties of random laser.

### 5.1 Calculation

For both scattering particles I have calculated the scattering cross section following equation (2.9). It was found that scattering cross section is $1.23 \times 10^{-15} \text{ m}^2$ and $2.86 \times 10^{-13} \text{ m}^2$ for large size and small size nano-particle respectively. Next, using equation (2.10) transport mean free path was calculated for different concentrations of both scatterers. Table (5.1) shows the change of $l_t$ with various concentrations. It

<table>
<thead>
<tr>
<th>Sample concentration</th>
<th>Particle size</th>
<th>Transport mean free path $(l_s \approx l_t)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$10^{10} \text{cm}^{-3}$</td>
<td>100 nm</td>
<td>650.24 nm</td>
</tr>
<tr>
<td>$10^{11} \text{cm}^{-3}$</td>
<td>100 nm</td>
<td>65.08 nm</td>
</tr>
<tr>
<td>$10^{12} \text{cm}^{-3}$</td>
<td>100 nm</td>
<td>6.51 nm</td>
</tr>
<tr>
<td>$10^{13} \text{cm}^{-3}$</td>
<td>100 nm</td>
<td>0.651 nm</td>
</tr>
<tr>
<td>$10^{9} \text{cm}^{-3}$</td>
<td>300 nm</td>
<td>8.87 nm</td>
</tr>
<tr>
<td>$10^{10} \text{cm}^{-3}$</td>
<td>300 nm</td>
<td>0.90 nm</td>
</tr>
<tr>
<td>$10^{11} \text{cm}^{-3}$</td>
<td>300 nm</td>
<td>0.09 nm</td>
</tr>
</tbody>
</table>

**Table 5.1**: Transport mean free path for different concentration of two different size nano-particle.
is clear from the above table that with increasing the particle concentration, transport mean free path decreases. Only the smaller size particle having concentration \(10^{10}\) cm\(^{-3}\) provide the \(l_t\) larger than pumping wavelength. This has an important impact on multiple scattering, as well as in coherent random lasing. This will be discussed in the following sections.

I have also calculated the spectrometer resolution by considering the wavelength difference. The grating period \(d = 416.5\) nm, the angle between grating and incident light \(\theta_i = 30^\circ\), camera pixel width \(P = 5.2\) \(\mu\)m and the distance between the camera and the grating \(L = 5.5\) cm and \(\theta = |\theta_1 - \theta_2|\). Following the figure 5.1(a) resolution can be calculated as follows

\[
\tan\left(\frac{\theta}{2}\right) = \frac{P}{2L} = \frac{5.2 \times 10^{-6} m}{2 \times 5.5 \times 10^{-2} m} = 4.73 \times 10^{-5}
\]

\[
\frac{\theta}{2} = 2.71 \times 10^{-3}
\]

\[
\therefore \theta = 5.42 \times 10^{-3}
\] (5.1)

**Figure 5.1:** (a) Geometry of spectrometer resolution calculation, (b) Variation of spectrometer resolution as a function of wavelength.
For two different wavelengths $\lambda_1$ and $\lambda_2$ we can write
\[
\sin \theta_1 = \frac{\lambda_1}{d} - \sin \theta_i \\
\sin \theta_2 = \frac{\lambda_2}{d} - \sin \theta_i
\]  
(5.2)

Thus, we know
\[
\theta = |\theta_1 - \theta_2| = \sin^{-1}\left(\frac{\lambda_1}{d} - \sin \theta_i\right) - \sin^{-1}\left(\frac{\lambda_2}{d} - \sin \theta_i\right).
\]  
(5.3)

Let us consider $\lambda_1 = \lambda$, $\lambda_2 = \lambda - \Delta \lambda$ and $(\frac{\lambda}{d} - \sin \theta_i) = A$, then equation (5.3) can be written as
\[
\sin^{-1}(A) - \sin^{-1}(A - \frac{\Delta \lambda}{d}) = \theta \\
\sin^{-1}(A - \frac{\Delta \lambda}{d}) = \sin^{-1}(A) - \theta \\
A - \frac{\Delta \lambda}{d} = \sin(\sin^{-1}(A) - \theta) \\
\Delta \lambda = Ad - d \sin(\sin^{-1}(A) - \theta) \\
\therefore \Delta \lambda = \left(\frac{\lambda}{d} - \sin \theta_i\right)d - d \sin(\sin^{-1}\left(\frac{\lambda}{d} - \sin \theta_i\right) - \theta)
\]  
(5.4)

Using equation (5.4), the spectrometer resolution calculated for the wavelength range of green light and found between $2.86 \times 10^{-2}$ nm to $1.95 \times 10^{-2}$ nm. Figure 5.1 shows the change of spectrometer resolution with wavelength. The resolution changes with increasing the wavelength but it's quite enough for measuring the sharp spectral peaks of random laser. Experimentally it has been found that random lasing peak widths are less than $0.2$ nm [5]. Thus, it can be said that our spectrometer is capable to measure the random laser spectra.

### 5.2 Random lasing and analysis

Random lasing was determined in both weak and strong scattering medium by varying the concentration of scattering centre. The dye concentration is fixed and the nano-particle concentration varies from $10^9$ cm$^{-3}$ to $10^{12}$ cm$^{-3}$ for average particle size 300 nm and from $10^{10}$ cm$^{-3}$ to $10^{13}$ cm$^{-3}$ for average particle size 100 nm. The random medium having concentration between $10^9$ cm$^{-3}$ to $10^{10}$ cm$^{-3}$ is the
weak scattering medium and concentration between $10^{11}$ cm$^{-3}$ to $10^{13}$ cm$^{-3}$ is the strong scattering medium. During the experiment, when the broad pump pulse is incident on the front surface of the sample the gain medium is excited optically. Light experiences multiple scattering and hence randomizes its directionality. Some of the scattered light leave the active media to the unpumped region and after a short period of time some of them return to the active volume again to be amplified more and provide positive feedback.

In weak scattering medium light can easily escape the active volume, but in the case of strong scattering medium, light experiences dense multiple scattering. The strong scattering medium forms a closed loop through recurrent process. Therefore, light stays a longer period of time in active medium and gets amplified more. Also, the path length is longer, but transport length is shorter in dense scattering media. When the light escapes the active volume in a short period of time in a diffusive way, the less cavities are formed or the recurrence probability becomes low. Consequently, the optical loss becomes high. While the pump energy is large enough, the gain becomes high, or in strong scattering volume, light is confined in a certain area for a longer time and scattered multiple times, thus experiencing large gain.

In multiple scattering region, the photon generation length is smaller than the path length, and as a result, the photon generation rate in this region is high. When the photon generation rate balances with the photon loss rate, a lasing threshold is build up in the amplified spontaneous emission (ASE) spectrum. At a low pump pulse intensity, the emission spectra exhibit only a broad (ASE). As the incident pulse intensity is increased, the spectrum becomes narrower, showing a small bandwidth. After a large pump pulse, the spectrum reaches the threshold where the loss is balanced with gain. Later on, when the pump intensity exceeds the threshold, i.e. photon generation rate is higher than the photon loss, the emission spectrum is broken up and drastic lasing peaks appear on top of the spectrum.

In our experiment, the pump pulse intensity ($\sim 800$ W/m$^2$) was constant and large enough to exceed the threshold and to produce lasing. Figure 5.2 shows the spectral emission images and the corresponding laser emission peaks with scatterers and without scatterers. Figure 5.2 (a) and (b) represent the spectral image of non-scattering and scattering medium, respectively. In the figure 5.2 (a) there is a large bright spot, but in figure 5.2 (b) there several bright spots. The bright spots indicate that cavities are formed and light interferes constructively. Corresponding to these
images, there is a large emission spectrum without peaks shown in figure 5.2 (c), but there are discrete peaks in figure 5.2 (d). The reason behind it is, that in an empty sample there is only gain medium, and when it’s pumped it emits ASE spectrum.

Figure 5.2: The spectral images and corresponding spectrum for a random medium without scatters and with scatters.

After performing the experiment, the effect of different concentration in random lasing is observed. For low concentration of small particle the $l_t$ is quite large, 650.24 nm, which is even larger than the pump pulse wavelength. But, for the high concentration it’s reduced to 0.651 nm. Large nano-particle provide short transport length at low concentration (8.87 nm) compare to the small particle. But, at high concentration it is considerably too small. This variation of transport length has a clear impact on random lasing.
On the contrary, in the strongly scattering media the transport length is small but photon generation length is large. This means that in strong active region light faces multiple scattering and randomizes its direction. Hence, the probability of light returning to its initial scattering point becomes high. Therefore, recurrent scattering occurs and the cavity formed through this process experiences low losses. Also, in the strong scattering volume, more cavities are formed and light is trapped inside the active region for a longer time and amplified more. Accordingly, the gain increases dramatically and threshold decreases rapidly. Thus, the threshold is low in strong scattering medium and the lasing occurs at low pump pulse energy. Figure 5.3

![Figure 5.3: The lasing emission spectrum for concentrations $10^{10}$ cm$^{-3}$, $10^{11}$ cm$^{-3}$, $10^{12}$ cm$^{-3}$ and $10^{13}$ cm$^{-3}$ of scattering particle size 100 nm.](image)

shows the lasing mode variation for different concentration of nano-particles having average scatterers diameter of 100 nm. Figure 5.3 (a) shows that on the top of the emission spectrum, there are some small emission peak for the concentration of $10^{10}$ cm$^{-3}$. With close observation of figure 5.3 (b), I found that the number of spectral
peaks increases with increasing the particle concentration. Similarly, figure 5.3 (c) and (d) exhibit more sharp and numerous lasing peaks for a high concentration of $10^{12}$ cm$^{-3}$ and $10^{13}$ cm$^{-3}$, respectively. This is because of large, low loss cavities at high concentrations. This variation in emission peaks in the spectrum occurs, because with increasing the particle concentration transport length decreases and approaches the pump wavelength. When it is approximately equal to the pump wavelength, the threshold intensity drops quickly. For concentration $10^{12}$ cm$^{-3}$ (100 nm) transport length (650.24 nm) is close to the incident wavelength (532 nm). If we relate the impact of this transport length on figure 5.3 (a), we see that lasing is just starting at this concentration. But, when transport mean free path becomes smaller than the pump wavelength, a drastic emission occurs rapidly. If we compare the transport length from table (6.1) to the spectrum of figure 5.3 (b, c, d) we find that for low transport length, below the pump wavelength, spectrum exhibits more peaks.

(a) (b)

(c) (d)

**Figure 5.4:** The lasing emission spectrum for concentrations $10^9$ cm$^{-3}$, $10^{10}$ cm$^{-3}$, $10^{11}$ cm$^{-3}$ and $10^{12}$ cm$^{-3}$ of scattering particle size 300 nm.
Next, we observed the lasing behaviour for different concentrations of large diameter (300 nm) nano-particle. Figure 5.4 (a)-(d) shows the emission spectrum of random laser having concentration $10^9$ cm$^{-3}$, $10^{10}$ cm$^{-3}$, $10^{11}$ cm$^{-3}$ and $10^{12}$ cm$^{-3}$, respectively. Where, spectrum (a) and (b) exhibit ASE with sharp lasing peaks on top of it. In comparison between spectrum (a) and (b), we see spectrum (b) show more sharp peaks than (a) because the transport length $l_t$ is almost ten times shorter than the first one. This indicate that more local low loss cavities are formed on (b) due to large concentration. Whereas, spectrum (c) and (d) shows a narrow sharp spectrum without any peaks. The fill factor of TiO$_2$ are $1.76 \times 10^{-3}$ and $1.76 \times 10^{-3}$ in the concentration $10^{12}$ cm$^{-3}$ and $10^{13}$ cm$^{-3}$, respectively, which is quite large. For such a large fill factor of TiO$_2$ the scatterers volume become much larger than the gain volume. When the pump light travels through the random medium the amplification does not become effective during propagation. As the number of cavities increases with increasing the particle density, the number of quasi degenerate modes also increases with it. Simultaneously, the lasing modes lead to an increase in the emission intensity, but there is a limit for maximum particle number density per unit volume, above which the lasing intensity decreases [21]. Thus, the particle number density exceeded the limit of maximum number density per unit volume in the concentration of $10^{12}$ cm$^{-3}$ and $10^{13}$ cm$^{-3}$. Consequently, there is no lasing taking place in such medium.

5.3 Coherence characterization

As the random medium provides both coherent and incoherent feedback, in this experimental work coherent properties of random lasing were measured. The measurements of coherence characteristics were performed with the grating interferometer. For a series of concentrations of two different size scattering particles, the maximum visibility and its full width half maximum (FWHM) were determined. The coherence between two points is determined from the visibility of interference fringe.

Table 5.2 shows the maximum visibility and its full width half maximum (FWHM) for different concentration of two scattering particles. The spatial coherence width is denoted by FWHM. From the table 5.2 its found that, the empty sample provides maximum visibility of 0.46 and its FWHM 7$\mu$m. Of course, the empty sample should be completely incoherent but it provides visibility, because of the Van Cittert-
<table>
<thead>
<tr>
<th>Sample concentration</th>
<th>Particle size</th>
<th>FWHM</th>
<th>Maximum visibility</th>
</tr>
</thead>
<tbody>
<tr>
<td>$10^{10} cm^{-3}$</td>
<td>100 nm</td>
<td>7.5 μm</td>
<td>0.692</td>
</tr>
<tr>
<td>$10^{11} cm^{-3}$</td>
<td>100 nm</td>
<td>12 μm</td>
<td>0.622</td>
</tr>
<tr>
<td>$10^{12} cm^{-3}$</td>
<td>100 nm</td>
<td>9 μm</td>
<td>0.659</td>
</tr>
<tr>
<td>$10^{13} cm^{-3}$</td>
<td>100 nm</td>
<td>13.8 μm</td>
<td>0.604</td>
</tr>
<tr>
<td>$10^{9} cm^{-3}$</td>
<td>300 nm</td>
<td>12 μm</td>
<td>0.639</td>
</tr>
<tr>
<td>$10^{10} cm^{-3}$</td>
<td>300 nm</td>
<td>10 μm</td>
<td>0.738</td>
</tr>
<tr>
<td>Empty sample</td>
<td>7 μm</td>
<td></td>
<td>0.46</td>
</tr>
</tbody>
</table>

Table 5.2: Maximum visibility for different concentration.

Zernike theorem. As a result, generated fringes that show spatial coherence. It is easier to understand the spatial coherence of random laser with comparing it to the spatial coherence of ASE. Also, random laser emission contains both lasing and ASE, therefore spatial coherence of RL is similar to empty sample [47].

It is clear from the table 5.2 that for the smaller size particle with increasing the concentration, the maximum visibility decreases, except for concentration $10^{13} cm^{-3}$, which exhibits larger visibility than its nearest neighbouring concentrations. Additionally, spatial coherence width increases with increasing the concentration, except for $10^{13} cm^{-3}$, and also it becomes wider than the empty sample. On the other hand, maximum visibility increases with increasing the concentration for large nanoparticle. But, FWHM decreases with increasing concentration although its larger than the empty sample.

In this experiment, visibility fluctuates although the pump pulse energy was constant (100 mJ) for all concentrations. This could be due to the wavelength sensitive interference. Even in all the cavities, light can not be confined at the same wavelength and can't interfere constructively. Furthermore, cavities in random medium are mobile and continuously change their configuration. We used magnetic stirring inside the sample to keep a constant density, but due to the random movement of particles, the number of cavities and its spatial structure also varied in the active volume, which also causes the fluctuation in the visibility. Moreover, continuous cavity mobility causes incomplete cavity loop and thus losses increase. Inside the incomplete cavity loop light does not interfere constructively. During the mobility of particle, the back scattering angle is changed which causes the fluctuation
Figure 5.5: Maximum visibility for different concentration.

of emission intensity. Figure 5.5 shows the visibility of different concentration of two types of scatters. It shows that the visibility also considerably high at different axial positions. Therefore, from experimental result and above discussion it can be concluded that the random lasing source is partially spatially coherent.
In summary, we have observed the spatial coherence properties of random laser using a new experimental method: double-grating interferometer. Our random disordered medium is rhodamine 6G doped TiO$_2$ nano-particles dissolved in methanol. A series of concentrations were prepared for two different size particles having a diameter of 100 nm and 300 nm. For the large scattering particle our concentrations were $10^9$ cm$^{-3}$ to $10^{12}$ cm$^{-3}$ but for small scattering particle it was $10^{10}$ cm$^{-3}$ to $10^{13}$ cm$^{-3}$. First, we have determined the lasing behaviour of disordered medium for all prepared concentrations. The pump source was a Q-switched Nd:YAG laser with constant pump intensity of 800 W/m$^2$. The pump intensity was high enough, above the lasing threshold, which causes the medium to lase. We have studied the lasing mode variation with fill factor of scatters, which is inversely related to transport or scattering mean free path, and particle diameter. It was observed that for smaller size scatterers, with increasing the particle concentration, lasing spectra shows more sharp peaks in it. In the case of larger particles, random media having concentrations $10^9$ cm$^{-3}$ and $10^{10}$ cm$^{-3}$ provide enormous lasing. But, above these concentrations emission spectra showed only a narrow ASE line. It was because of particle number exceed the maximum density per unit volume. Furthermore, we calculated the spectrometer resolution which implies that our sensor can detect the very sharp lasing peaks.

After observing the active random medium which is showing good lasing spectra, we determined the spatial coherence of this lasing phenomenon. We used the grating interferometer method to determine the visibility. We also determined the visibility of empty sample, without scatters, so that we can compare the variation
of visibilities. From these values we came to the conclusion that our random laser source was partially spatially coherent. Our result is quite important in the application of random laser. We suggest that the developed random laser is useful in low coherence and high intensity cases where conventional laser is not suitable.


This matlab code has written by Matias Koivurova.

```matlab
name='10.10.300.2.avi';
height=7;
workingDir = tempname;
mkdir(workingDir)
mkdir(workingDir,'images')
RLVideo = VideoReader(name);
ii = 1;
while hasFrame(RLVideo)
    img = readFrame(RLVideo);
    filename = [sprintf('%03d',ii) '.jpg'];
    fullname = fullfile(workingDir,'images',filename);
    imwrite(img,fullname)% Write out to a JPEG file (img1.jpg, img2.jpg, etc.)
    ii = ii+1;
end
imageNames = dir(fullfile(workingDir,'images','*.jpg'));
imageNames = {imageNames.name}';
for ii = 1:length(imageNames)
    img = imread(fullfile(workingDir,'images',imageNames{ii}));
    A(ii,:)=(img(height,:,1));
end
w=(547+0.0285714*[1:1280]);
figure;
imagesc(A);
colorbar;
figure;
plot(w,A(1,:))
```
The basic matlab code for visibility measurement has written by Matias Koivurova but I have modified it to analyse the data.

```matlab
function [Vis ]= visibility_2(Name,height)
tic
%the number of pictures in the file
numFiles=100;
%height from where you are taking the cross section
%height=100;
%number of sampling cross sections
samples=1;
%distance between each sample in pixels
distance=10;
%noise reduction (no need to change this)
noise=2;
t=cell(samples,numFiles);
% Load pictures
for fileNum=1:numFiles;
    fileName=sprintf('C:/Users/TaHeR KhaN/Documents/MATLAB/18.3.16 Coherence data/10-10-300/average pihole size 10-10-300/img%s%d.png',Name,fileNum);
a=imread(fileName);
    for c=1:samples;
        t{c,fileNum}=a(height+(c-1)*distance,:);
    end
end
```
%---------------------------------------% Computation %
for c=1:samples;
    V=zeros(numFiles,1280);
%---------------------------------------% compute visibility %
for k=1:numFiles;
    img=im2double(t{c,k}-noise);
    [Maxima,MaxIdx]=findpeaks(img);
    DataInv=max(img)-img;
    [Minima,MinIdx]=findpeaks(DataInv);
    if length(Maxima)<length(Minima);
        l=length(Maxima);
        h=Maxima-(max(img)-Minima(1,1:l));
        p=Maxima+(max(img)-Minima(1,1:l));
        V(k,MaxIdx)=(h./p);
    else
        l=length(Minima);
        h=Maxima(1,1:l)-(max(img)-Minima);
        p=Maxima(1,1:l)+(max(img)-Minima);
        V(k,MaxIdx(1,1:l))=(h./p);
    end
end
Vis=zeros(1,numFiles);
for k=1:numFiles;
    a=max(V(k,:));
    Vis(1,k)=a;
end
%Vis900C=Vis;
%Vis900A=Vis;
%Vis900B=Vis;
%Vis500B=Vis;
%Vis500C=Vis;
%Vis100A=Vis;
%Vis100B=Vis;
%Vis100C=Vis;
%figure;
%imagesc(V')
toc
h=[100 500 900];
Name=char('A', 'B', 'C');
k=1;
for i1=1:3
    for j=h
        Vis(k,:)= visibility_2(Name(i1),j);
        k=k+1;
    end;
end;
Avg =mean(Vis(1:end,:));
Max=max(Avg);
plot(Avg);

Avg2=Avg-min(Avg);
Max2=max(Avg2);
figure; plot(Avg2);