Analysis of mid-infrared lasing in active random media

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Abstract: Lasing behaviour of 2-dimensional active random structures, designed to work in the Mid-IR region, has been investigated at different input powers by varying the amount of scattering intensity. A Monte Carlo based simulation tool has been developed including a model to manage the optical amplification. The analysis of photon travel distance has been considered to show the random lasing behaviour with particular attention on lasing threshold at different scattering intensity. The simulated results are in agreement with experiments.

OCIS codes: (250.5960) Semiconductor lasers; (140.3380) Laser materials; (290.4210) Multiple scattering.

References and links

1. F. Pratesi, M. Burresi, F. Riboli, K. Vynck, and D. S. Wiersma "Disordered photonic structures for light harvesting in solar cells," Opt. Express **21**(S3), A460–A468 (2013).
2. B. Redding, M. A. Choma, and Hu Cao, "Speckle-free laser imaging using random laser illumination," Nature Photon. **6**, 355–359 (2012).
3. D. S. Wiersma, P. Bartolini, A. Lagendijk, and R. Righini, "Localization of light in a disordered medium," Nature **390**, 671–673 (1997).
4. M. Segev, Y. Silberberg, and D. N. Christodoulides "Anderson lacalization of light," Nature Photon. **7**, 197–204 (2013).
5. A. L. Burin, M. A. Ratner, H. Cao, and S. H. Chang, "Random laser in one dimension," Phys. Rev. Lett. **88**, 093904 (2002).
6. V. S. Letokhov, "Light generation by a scattering medium with a negative resonant absorption," Sov. Phys. JETP **16**, 835–840 (1968).
7. N. M. Lawandy, R. M. Balachandran, A. S. L. Gomes, and E. Sauvain, "Laser action in strongly scattering media," Nature **368**, 436438 (1994).
8. D. S. Wiersma, "The physics and applications of random laser," Nature Phys. **4**, 359–367 (2008).
9. K. L. van der Molen, R. W. Tjerkstra, A. P. Mosk, and A. Lagendijk, "Spatial extent of random laser modes," Phys. Rev. Lett. **98**, 143901 (2007).
10. P. Sebbah and C. Vanneste, "Random laser in the localized regime," Phys. Rev. B **66**, 144202 (2002).
11. J. Andreasen, A. A. Asatryan, L. C. Botten, M. A. Byrne, H. Cao, L. Ge, L. Labont, P. Sebbah, A. D. Stone, H. E. Treci, and C. Vanneste, "Modes of random lasers," Adv. Opt. Photon. **3**(1), 88–127 (2011).
12. J. Fallert, R. J. B. Dietz, J. Sartor, D. Schneider, C. Klingshirn, and H. Kalt, "Co-existence of strongly and weakly localized random laser modes," Nature Photon. **3**, 279–282 (2009).
13. H. K. Liang, B. Meng, G. Liang, J. Tao, Y. Chong, Q. J. Wang, and Y. Zhang, "Electrically pumped mid-infrared random lasers," Adv. Mater. **25**(47), 6859–6863 (2013).
14. C. Molardi, X. Yu, H. Liang, Y. Zhang, A. Cucinotta, and S. Selleri, "Modal analysis in 2D media with variable disorder," Opt. Express **23**, 3681-3689 (2015).

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15. R. Uppu and S. Mujumdar, "Dependence of the Gaussian-Lévy transition on the disorder strength in random lasers" Phys. Rev. A **87**, 013822 (2013).
16. J. Faist, F. Capasso, D.L. Sivco, C. Sirtori, A.L. Hutchinson, and A.Y. Cho, "Quantum cascade laser," Science **264** (1994) 553–556.
17. L. C. Henyey and J. L. Greenstein, "Diffuse radiation in the galaxy," Astrophys. J. **93**, 70–83 (1941)
18. E. Zielinski, H. Schweizer, K. Streubel, H. Eisele, and G. Weimann, "Excitonic transitions and exciton damping processes in InGaAs/InP" J. Appl. Phys. **59**(6), 21962204 (1986).
19. J. Faist, F. Capasso, C. Sirtori, D. Sivco, and A. Cho, *Intersubband Transitions in Quantum Wells: Physics and Device Applications II* (Academic, 2000, Vol. 66).
20. S. Mujumdar, M. Ricci, R. Torre, and D. S. Wiersma. "Amplified extended modes in random lasers," Phys. Rev. Lett. **93**, 053903 (2004).

1. Introduction

In recent years the behaviour of light in photonic disordered materials has tickled the interest of scientific community. The applications, where the mesoscopic properties of light can be exploited, are becoming more and more tangible, ranging from energy efficiency research to imaging and sensing field [1, 2]. Inside a structure characterized by a certain level of disorder, the light experiences the physical phenomenon of multiple scattering. The interaction with scattering centres randomizes light diffusion, making the photon travel similar to a random walk [3]. The scattering intensity is fundamental to explain the light transport. Increasing its contribution, the transport changes from ballistic to diffusive, reaching a localized regime when the scattering is enough intense [4]. Localization can be described as the development of close channels where light is trapped by the feedback due to the scattering [5]. This situation is referred as strong localization and can be reached when the modified Ioffe-Regel criterion $kl \leq 1$ is satisfied, where k is the wave number and l is the photon mean free path [3]. Backscattering is a second mechanism identified to explain localized regime. Its action is considered a transition between diffusion and localization, therefore it is referred as weak localization [4].

As theoretically predicted by Letokhov [6] and later demonstrated by Lawandy [7], the disorder combined with the amplification given by an active material can be exploited to create a new class of lasers with particular properties. Inside a disordered active material the light, forced to travel through longer paths, can reach the lasing threshold showing an emission spectrum characterized by narrow peaks apparently not correlated with each others [8]. Comparing to a traditional laser the feedback mechanism is due to the scattering and not to the action of an optical cavity. This class of laser is normally referred as random laser. Random lasing is a complex phenomenon, which can not be explained with the only help of localization theory. In a strong scattering regime the interference can give birth to amplified optical modes characterized by a high Q-factor and spatial separation, namely localized modes. In a low scattering regime the light is diffused all over the material and lasing peaks can be explained as the amplification of the photons which experience very long paths. Such events can be improperly referred as extended open modes [9]. While there is a clear relation emerging from the comparison between leaky modes and amplified modes in localized regime [10], such relation appears very weak in diffusive regime. A rigorous explanation of this behaviour has been given in [11], where it is evidenced the substantial difference between passive and active modes. While the passive modes can be described as poles of a unitary scattering matrix, in an active system the gain makes the scattering matrix no more unitary. A direct consequence is the onset of a shift in frequency and field distribution between modes of passive system and amplified modes. For high Q-factor modes in localized regime this shift is minimum. In contrast in a weak scattering regime such shift leads to a complete inconsistency between leaky and amplified modes. Experiments show that different optical modes, open and localized, can co-exist and interact with the active medium competing for the gain [12]. In this scenario a key factor to describe the behaviour of active disordered structures in presence of a moderate scattering is related to the

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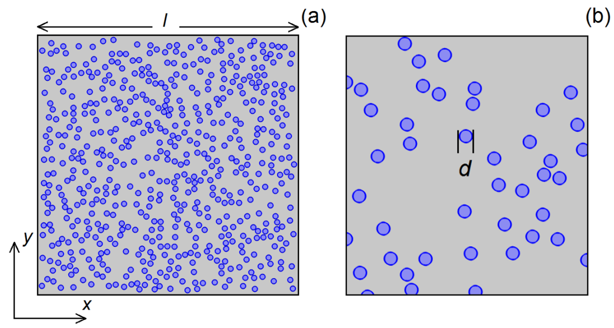

Fig. 1. (a) Schematic of a disordered planar slab. The slab is a square with side l equal to 150 µm. (b) The scattering is introduced carving a pattern of circular holes with diameter d equal to 3 µm.

study of the photon travel distance inside the material.

In this work a planar structure, obtained carving a random pattern of holes in a Quantum Cascade Laser (QCL) substrate and designed to lase in Mid-IR region [13], has been investigated. A simulator based on Monte Carlo (MC) method, modified to include a pumping system, has been developed to calculate the travel distance of photon inside the media [14]. Structures with different amount of scattering has been simulated, progressively increasing the input power.

2. 2D disordered structure

The structure under examination, shown in Fig. 1(a), is square planar slab obtained in a QCL heterostructure made with a compound of $In_{0.53}Ga_{0.47}As$ / $In_{0.52}Al_{0.48}As$. The QCL stack contains 35 active/injector stages. The refractive index n_s of the substrate is equal to 3.35 . The QCL has been supposed emitting in a band around 10 µm, in the middle infrared region. The square side length l is equal to 150 µm and thickness t is equal to 2.2 µm. Despite the slab has a well-defined thickness, the structure has been considered 2-dimensional for two reasons. The first is related to the large ratio between the square side l and the thickness t, and the second is related to the properties of QCL substrate which mainly supports transverse magnetic modes [16]. Inside this slab several circular holes, which act as centres of scattering, have been carved. The holes have identical diameter of 3 µm, Fig. 1(b), and are located with a random strategy such as their position is a random variable with a uniform distribution. To avoid overlaps a guard distance of 150 nm has been set between the holes. As a measure of the scattering, the filling factor (FF) has been chosen as core parameter. This parameter is defined as the total area of the holes divided by the total area of the square surface. Finally it has been assumed that the slab is surrounded by a passive material, precisely air.

3. Monte Carlo method in active medium

MC method is commonly used in bio-imaging and microscopy to model photons propagation in scattering media. With the advantage to be simple and computationally not expensive, this method assumes that photons act as particles interacting with the scattering media with a sequence of random and mutually independent events. Consequently MC strategy ignores the light interference. Even though a correct numerical description of random lasing phenomenon should be carried out exploiting the solution of Maxwell equations, the approximation given by the use of MC method can be considered acceptable in the weak scattering region [15]. Furthermore MC makes available the statistics of light diffusion inside the media and makes possible

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the evaluation of disordered systems in function of filling factor, which is a global parameter independent of a particular realization of random pattern. This fact can be successfully used in the preliminary step of random laser design. Normally MC method is applied in passive media where the photons action is independent. To investigate the behaviour of the disordered active structure shown in Fig. 1(a), a software based on MC method has been implemented including a pumping scheme and a gain model. The photon energy has been chosen as the main physical quantity to describe the evolution of the system. The structure has been divided in a discrete grid and every block of the grid has been described as a bucket of energy which can be empty in the case of pump lack or full in case of saturation. Every photons packet, passing in a particular bucket region, interacts with it absorbing energy via stimulated emission. The amount of energy in the bucket is also decreased by spontaneous emission which is the starting spark of random lasing phenomenon in the structure proposed. The equations of spontaneous emission, stimulated emission and absorption have been written to show the energy:

$$\Delta E_{sp} = E_b \left(1 - e^{A\Delta t}\right), \tag{1}$$

$$\Delta E_{th} = E_{pk} \left(1 - e^{-\Sigma \Delta l}\right), \tag{2}$$

$$\Delta E_{st} = E_{pk} \left(e^{\sigma \frac{E_b}{h\nu V_b} \Delta l} - 1\right), \tag{3}$$

where E_{sp}, E_{st} are the energy released by the upper state through spontaneous emission and stimulated emission respectively, E_{th} is the energy absorbed by the structure and dissipated through thermal effects. E_b and E_{pk} are the energy of a generic discrete bucket and the energy of a generic photon packet running in the media. The parameters A, Σ and σ are the rate of the spontaneous emission, the absorption coefficient and the emission cross section respectively, h is the Planck's constant, ν is the working frequency and V_b is the volume of a single cell. Δt and Δl are respectively the time step and the distance step emerging from the discretization. The aim of the bucket division is to better describe the interaction of photons packets with matter and their competition to obtain amplification, assuming that carriers diffusion inside the matter is much slower respect the photons diffusion. To model the disordered QCL substrate some simplifications have been made. The system has been assumed as a three-level system, operating at a cryogenic temperature of 78 K. Under this realistic consideration, photon re-absorption can be neglected and the Eq. (2) can be written considering only the thermal losses. The population inversion is due by the electrons injected in QCL stack through a pumping mechanism which fills equally all the buckets with an energy $\Delta E_b = P\Delta t$, where P the pump power. The gain cross section σ includes the contribution of all the stages in the cascade stack. The spontaneous emission has been modelled randomly choosing a number M of emission cells every time step. The scattering action has been concentrated on scattering centres and the deviation angle statistic has been calculated using the Mie theory with the approximation given by the Henyey-Greenstein function [17].

4. Simulations and results

The structure has been simulated using three different values of filling factor: 0.30, 0.25 and 0.18, as in [13]. These *FF* values suggest a diffusive regime, as shown in the modes evolution depicted in Fig. 2 . Furthermore referring to these FFs, the calculated Q-factor is low and the Ioffe-Regel criterion is not fulfilled [14]. As a consequence the use of MC method, which neglects the interference effect, appears justified. For every value of filling factor, ten different realizations of disorder have been considered and the statistical results collected from the simulations have been averaged. The simulations have been performed progressively increasing the input power from 0 to 40 W, which is equivalent to a current density range from 0 to

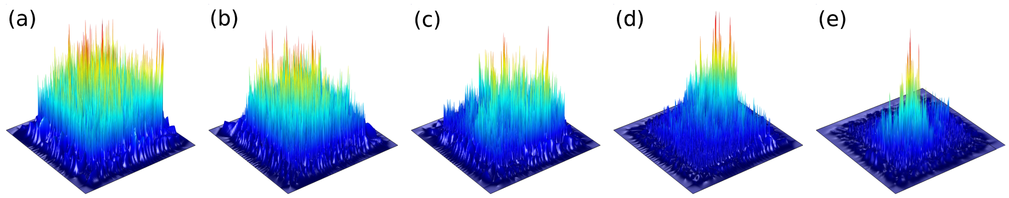

Fig. 2. Normalized electric field module at different FFs: (a) 0.18, (b) 0.25, (c) 0.30, (d) 0.36, (e) 0.42. Localization onset arises for FF greater than 0.4 .

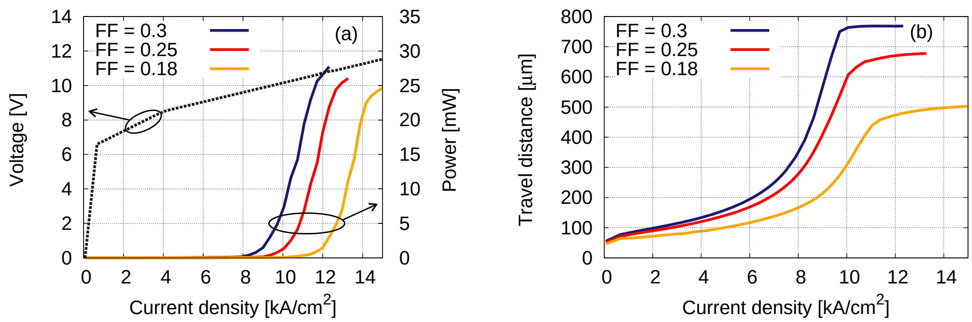

Fig. 3. (a) At left: voltage/current characteristic used for the simulation. At right: calculated Output power vs Current density at different values of filling factor FF. (b) Calculated average photon travel distance vs current density.

15 kA cm^{-2} according to the voltage/current characteristic shown in Fig. 3(a) and considering a pulsed operation with pulse length of 200 ns and repetition rate of 10 kHz. To model the QCL substrate, the parameters in Eqs. (1)-(3) have been set to the following values: Σ equal to 1.6×10^5 m^{-1} [18], A equal to 1.7×10^7 Hz and σ equal to 4.5×10^{-21} m^2 [19]. Figure 3(a) shows the output power versus the current density, it is possible to notice the dependence of the threshold, which requires higher current with the decrease of the filling factor. Furthermore the transition from non emission to emission region is sharper with respect of higher filling factor. The results obtained by simulation are comparable with experimental results shown in [13]. To better understand the process of amplification which occurs inside the slab, the software has been exploited to record the travel distance of each photons packet form its emission to its life termination, which can be a total absorption or an emission outside the slab. The average travel distance is shown in Fig. 3(b). For small current density values, far from the threshold, the absorption dominates over the stimulated emission and the weak spontaneous emission can not be amplified. The average path length remains low, with a value comparable with slab side, and exhibits a modest slope. Approaching the threshold the average travel length increases quickly reaching a saturation values just beyond the threshold point. The saturation length can be explained with the equilibrium between the amplified stimulated emission and the output power due to the slab size , while the transition zone before the threshold reflects the gain coefficient trend which locally can become positive with respect to the dynamic of packets diffusion. Such transition area is larger for smaller value of filling factor, justifying the smoother threshold.

The issue emerging from the previous considerations is related to the relative amount of stimulated emission with respect to non-amplified spontaneous emission of the output. Following

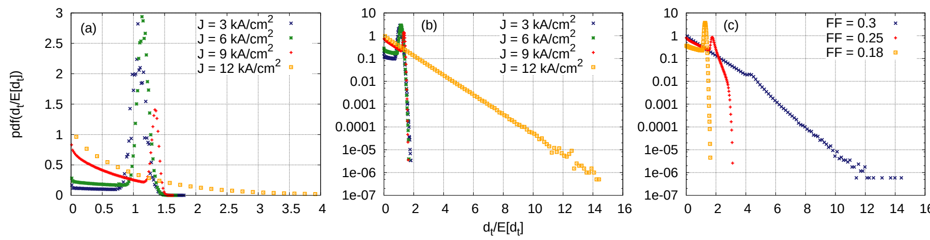

Fig. 4. Probability density function (pdf) of the travel length d_t normalized to its average $E[d_t]$ at FF equal to 0.3, written in linear scale (a) and in logarithmic scale (b), calculated at different values of current density J. (c) pdf calculated for various values of FF at $J = 10\,\mathrm{kA\,cm^{-2}}$, shown in logarithmic scale.

the idea which explains random lasing in weak scattering as a contribution of open modes, i.e. the development of photon packets with a path enough long to onset lasing phenomenon [20], the MC software has been exploited to investigate the statistic of photon packets. The results, shown in Fig. 4(a), depict the statistic (probability density function - pdf) of the travel length d_t normalized to its average value $E[d_t]$ in the case of $FF = 0.3$. Four different input current densities have been chosen to exhibit the statistical evolution of the system: 3, 6, 9 and $12\,\mathrm{kA\,cm^{-2}}$. The first and the second value are in a region far from the threshold, the third is in threshold proximity and the latter is in the emission region. Analysing the plot, it is possible to notice that in the lossy region the majority of the photon packets is characterized by a travel distance almost equal to the average distance, with a distribution that can be roughly compared to a narrow gaussian. In the emission region instead the pdf is clearly described by a negative exponential distribution. The transition region near the left side of the threshold shows a distribution which is a composition of both these behaviours, revealing an evolution from gaussian to exponential. This scenario is in agreement with the open modes interpretation of the diffusive regime, which describes a system dominated by spontaneous emission at low pumping level, and a system where long path packets can gain enough to lase at a robust pump level. The onset of long path packets is revealed by the transition to negative exponential distribution, as it is shown in Fig. 4(b), where the pdf is plotted in logarithmic scale. A relevant fraction of packets has a travel length several times longer than the average.

The travel length statistic dependence on the scattering straight is also relevant. As previously remarked, the filling factor influences the average travel length but the key role is played in relation with the pumping mechanism and the dynamic of the photon diffusion. Figure 4(c) shows the distribution of the travel length at different filling factor values, calculated at the input of $10\,\mathrm{kA\,cm^{-2}}$. Such location is interesting because it groups the statistics at the threshold transition increasing the scattering impact. In the case of $FF = 0.3$ the system is just beyond the threshold revealing the exponential distribution, while at $FF = 0.18$ the distribution is roughly gaussian. The case of $FF = 0.25$ is situated on the threshold transition while the pdf is evolving to exponential shape. These plots clearly explain the sensibility of the investigated planar slab to the change of filling factor. A proper design of the filling factor can, not only tailors the threshold but also the quality of the emission.

5. Conclusions

In this contribution a scattering optical planar structure built on a QCL active substrate with a randomly distributed pattern of holes and designed to lase in the Mid-IR region has been investigated. It has been assumed that such system is working in a weak scattering region, and the

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open modes interpretation of light diffusion has been selected to describe the system behaviour. A set of simulations, using a software based on a MC method, has been performed varying the input current density and the scattering amount, set through the filling factor parameter. The software has been implemented combining the MC method with a simple but complete model which includes the pumping system and the gain with particular attention to consider the local effects due to the resource competition. The focus has been given to the analysis of photon packets travel distance in order to explain the emission behaviour. Results shown that the emission threshold is correlated to the evolution of the travel length statistics from a gaussian-like to an exponential distribution. This fact justifies the amplified long path packets interpretation to explain lasing emission, dominated by the stimulated emission. Furthermore the emission characteristics calculated with the application of the modified MC method are in good agreement with experiments. This suggests that the simple and resource-inexpensive model proposed can be successfully applied to study and design a random system in regime of weak scattering.

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