Front. Optoelectron. China 2008, 1(1–2): 95–100
DOI 10.1007/s12200-008-0023-3

RESEARCH ARTICLE

Incoherent radiation of amplifying random media

Yunxia YE (✉)[1], Dianyuan FAN[2]

1 College of Mechanical Engineering, Jiangsu University, Zhenjiang 212013, China
2 Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, Shanghai 201800, China

Abstract Incoherent radiation of amplifying random media was investigated using Monte Carlo simulation and the characteristics of random laser were observed. The entire emission spectra of the random media would become narrow abruptly when pumping energies exceeded certain threshold values, and as the pumping energies further increased, distinct sharp peaks would emerge on the spectrum background. The intensity of a certain spatial point within the scattering media was contributed from emissions of many frequencies. The intensity of a single frequency of the entire spectrum was contributed with emissions from wide spatial location and angle ranges. It has been pointed out that the incoherent radiation of amplifying random media is essentially different from amplified spontaneous emission without any feedback as well as conventional resonant-feedback laser. The explanation on distinct sharp peaks is that rare photons have experienced rather more scattering events and walked longer paths in amplifying random media, which accumulates more gain for these photons.

Keywords random laser, incoherent radiation, Monte Carlo, scatter

1 Introduction

In the late 1960s, Letokhov predicted that laserlike emission would take place in amplifying random media by solving the diffusive equation with gain terms [1,2]. Later, it has been experimentally observed that emission spectra becomes narrow abruptly above certain thresholds in many kinds of amplifying random media, such as solid powder, dye scatterer systems and semiconductor powder [3–6]. The phenomenon was called "random laser". Thereafter, laserlike emission in amplifying random media became a hot topic for researchers. Now, there are two explanations on random lasers: resonant emission [7] and amplified nonresonant emission [4,8], using several research methods such as solving diffusive equations with gain terms [1,2], solving Maxwell equations using finite-difference time-domain (FDTD) method [7,9] and Monte Carlo simulation method [8,10]. Among those research methods, the Monte Carlo method has been used to trace photons in random media and then successfully explained the threshold and spectrum-narrowing phenomena with nonresonant emission. In this article, Monte Carlo method is used to simulate the nonresonant emission in amplifying random media. The entire spectra, spatial location and angle distributions of emission intensities have been calculated for different excitation energies. It has also been observed that the full width at half maximum (FWHM) of the emission spectra becomes narrow abruptly when excitation energies exceed certain thresholds and when these energies increase further, discrete sharp peaks will appear on smooth spectrum background. The spectra at certain spatial points, the spatial location and angle distribution of certain single frequency have been calculated too, which exhibit the characteristics of random laser from a new point of view.

2 Model

The system under study consists of an organic dye solution suspended with spherical scatterers homogeneously, and a pumping Gaussian light beam is normally incident on it. In the simulation, the particle number density N_s is 1.4×10^{19} m^{-3} and the dye concentration C is 3×10^{23} m^{-3}. The particle radius R is 60 nm. The refractive indexes of particle and solution are n_2 and n_1, respectively taken to be 1.9 and 1.4. To eliminate the influences of surface reflection on the photon propagating process in the random media, the surrounding refractive index n_0 is set equal to n_1.

Translated from *Chinese Journal of Lasers*, 2007, 34(3): 364–369 [译自: 中国激光]

E-mail: yx_ye@163.com

Sample thickness is designed to be 1 cm and its transverse dimension is much larger than the incident pumping beam radius r_{inc} which equals to 50 μm, so the influences of side faces of the random system on photon transport can be ignored. Pumping light has a wavelength of λ_{p} 532 nm and is assumed to work in single-pulse mode. Pulse width τ is far smaller than laser upper level lifetime τ_0. This assumption of $\tau \ll \tau_0$ is easily achievable in practice. The simulations are carried out for pumping energies E_{inc} of 0.5, 1.0, 1.5, 2.3 and 3.0 μJ. Here, dye is assumed as a standard four-level system. Thus, the population only distributes on the ground level and laser upper level. The population densities on these two levels are respectively N_0 and N_2, and $N_0 + N_2 = C$. Emission photons are collected from incident face.

The simulation is formed in two steps: pumping stage and emission stage. During pumping stage, the decrement of population on laser upper level is ignored because $\tau \ll \tau_0$. The absorption cross sections $C_{\mathrm{abs}}(\lambda)$, emission cross sections $C_{\mathrm{emi}}(\lambda)$ and emission spectrum of dye molecules are adapted from Ref. [11]. The average distance l between two scattering events is taken as $-l_{\mathrm{tr}}\ln(\xi)$, where ξ is a uniform variate in (0,1) and l_{tr} is transport mean free path which approximately equals to l_{s} because anisotropy factors are very small for the scatters used in this article [12]. l_{s} is scattering mean free path and taken as

$$l_{\mathrm{s}} = 1/N_{\mathrm{s}}C_{\mathrm{sca}}, \tag{1}$$

where scattering cross sections C_{sca} can be calculated according to Mie's theory [13]. The scattering angles θ after every scattering event is also calculated through Mie's theory and the azimuth angle ϕ evenly picked up between 0 and 2π.

During the pumping stage, the single shot excitation energies are divided into N_{p} energy packets, so the initial weight p of every packet is:

$$p = E_{\mathrm{inc}}\lambda_{\mathrm{p}}/hcN_{\mathrm{p}}, \tag{2}$$

where h and c are respectively Planck constant and velocity of light. Choosing N_{p} must consider both the precision and the cost time of calculation. Here, N_{p} is taken to be 10^6 and the calculation time is about 2 hours. The initial positions of energy packets are sampled according to Gaussian distribution and the initial incident angle is 0°. Then the pumping photons walk randomly in the media, accompanied with their absorption by the dye, so the photon weight p after each step must be updated as

$$p = p_0 \exp\left(-C_{\mathrm{pabs}}N_0(x,y,z)l\right), \tag{3}$$

where C_{pabs} is the absorption cross section at pumping wavelength and p_0 the photon weight before stepping. Simultaneously, the local population density $N_0(x,y,z)$ and $N_2(x,y,z)$ should be updated accordingly. So after excitation, a distribution of population density $N_2(x,y,z)$ of laser upper level is obtained, which acts as the input parameter of the next simulation stage.

During the emission stage, the position and associated wavelength λ of each emission photon are picked up according to the gain distribution constructed in the pumping stage and the normalized fluorescence spectrum of dye molecules. The initial scattering and azimuth angle are respectively sampled from 0–π and 0–2π uniformly. Between two scattering events, the photon undergoes losses by reabsorption and gains by stimulated emission. So the photon weight p must be updated as

$$p = p_0 \exp(-C_{\mathrm{abs}}N_0(x,y,z)l + C_{\mathrm{emi}}N_2(x,y,z)l). \tag{4}$$

The local population densities at ground and excited level should also be updated accordingly.

3 Results

In Figs. 1(a) and 1(b), the spontaneous emission spectrum and the spectra for different pumping energies: 0.5, 1.0, 1.5, 2.3 and 3.0 μJ are plotted. As shown, when pumping energy is low, only amplified spontaneous emission can be observed, and the intensity spatial distribution of which exhibits as a Gaussian distribution which is close to the pumping intensity distribution. When pumping energy increases further, the entire emission spectrum will become narrow. After a certain threshold, the spectra will narrow abruptly and a structure with several distinct sharp peaks on top of the smooth background appears. With the further increase of the excitation energy, the intensity and number of distinct sharp peaks increase accordingly. Figures 1(c) and 1(d) show the changes of FWHM and peak intensity with pumping energy, where FWHM and peak intensity are taken according to the inner envelope of the spectrum curve. From Fig. 1, it is found that for the random system in this article, the threshold E_{th} is about 1.0 μJ, and the emission spectrum and peak intensity change abruptly when $E_{\mathrm{inc}} \geqslant E_{\mathrm{th}}$.

From Fig. 2 it is shown that when $E_{\mathrm{inc}} < E_{\mathrm{th}}$, the spatial location and angle distribution of emission spectrum is almost smooth only with minor energy peaks on the top of the background spectrum. When $E_{\mathrm{inc}} > E_{\mathrm{th}}$, there emerges more energy peaks with stronger intensities. Figure 3 shows the spectra at certain spatial locations in the random media and it is found that the emission at local position is contributed from a wide spectrum range.

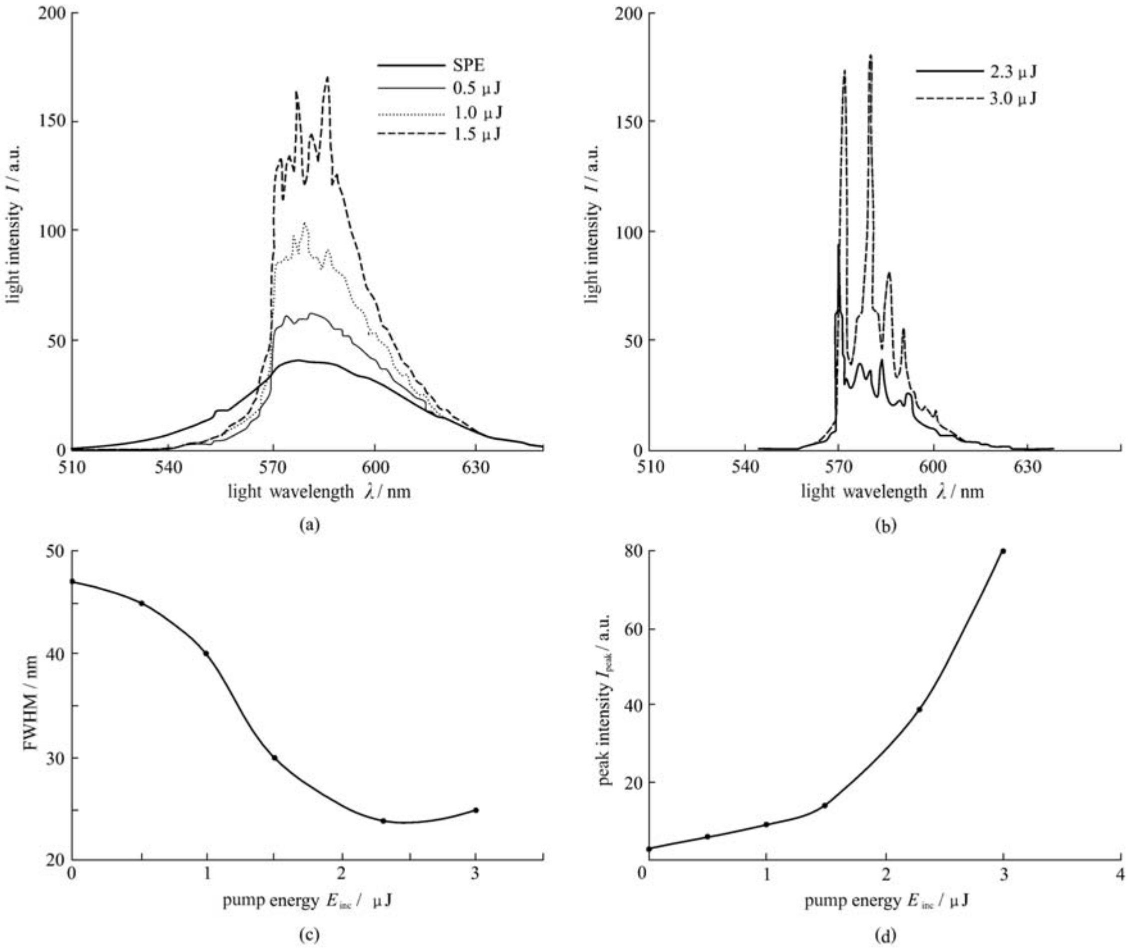

Fig. 1 Calculated emission spectra (a), (b), FWHM (c) and peak energies (d) at different pumping energies

4 Discussion on incoherent emission of amplified random media

4.1 Characteristics of incoherent random laser

Ignoring all losses except for the absorption by population on the ground level, the emission photon weight follows the law below:

$$p(\lambda)=p_0(\lambda)\exp\left(\int[-C_{\text{abs}}(\lambda)N_0(x,y,z)+C_{\text{emi}}(\lambda)N_2(x,y,z)]\mathrm{d}l\right). \quad (5)$$

From the above relationship, it can be concluded that the increasing excitation energy E_{inc} will make the emission within the gain line width amplified and the spectrum narrowed in three ways: 1) Increasing E_{inc}, the population density on the laser upper level increases and then the term of $C_{\text{emi}}(\lambda)N_2$ increases. Furthermore, the emission at the wavelength with larger value of $C_{\text{emi}}(\lambda)$ has the priority to be amplified; 2) Increasing E_{inc}, N_2 increases and N_0 decreases accordingly, so the term of $C_{\text{abs}}N_0$ decreases; 3) Increasing E_{inc} makes photons experience more gain and less loss, and then by combining the random scattering by particles, the resident time and walking distance of photons in the amplifying media will be prolonged, so the photon weight will be amplified further. This mechanism is completely similar to the conventional laser. When the cavity loss decreases, the feedback produced by cavity mirrors prolongs the resident time and walking path of the photon in the active media forms positive feedback. Therefore, the scattering feedback supplied by scatterers makes the emission of the amplifying random material different from common amplified spontaneous emission (ASE) without any feedback, and it is also the essential reason for the threshold behavior of nonresonant emission in amplifying random media [14].

On the other hand, emission from amplifying random media is also different from conventional laser although they both exhibit a threshold behavior. The function of scatterers is similar to that of cavity mirrors; however, the feedback they supply is random, so they cannot choose a specific light frequency or orientation. As shown in Figs. 2(c) and 2(d), many sharp peaks will emerge at different locations or in different directions in random

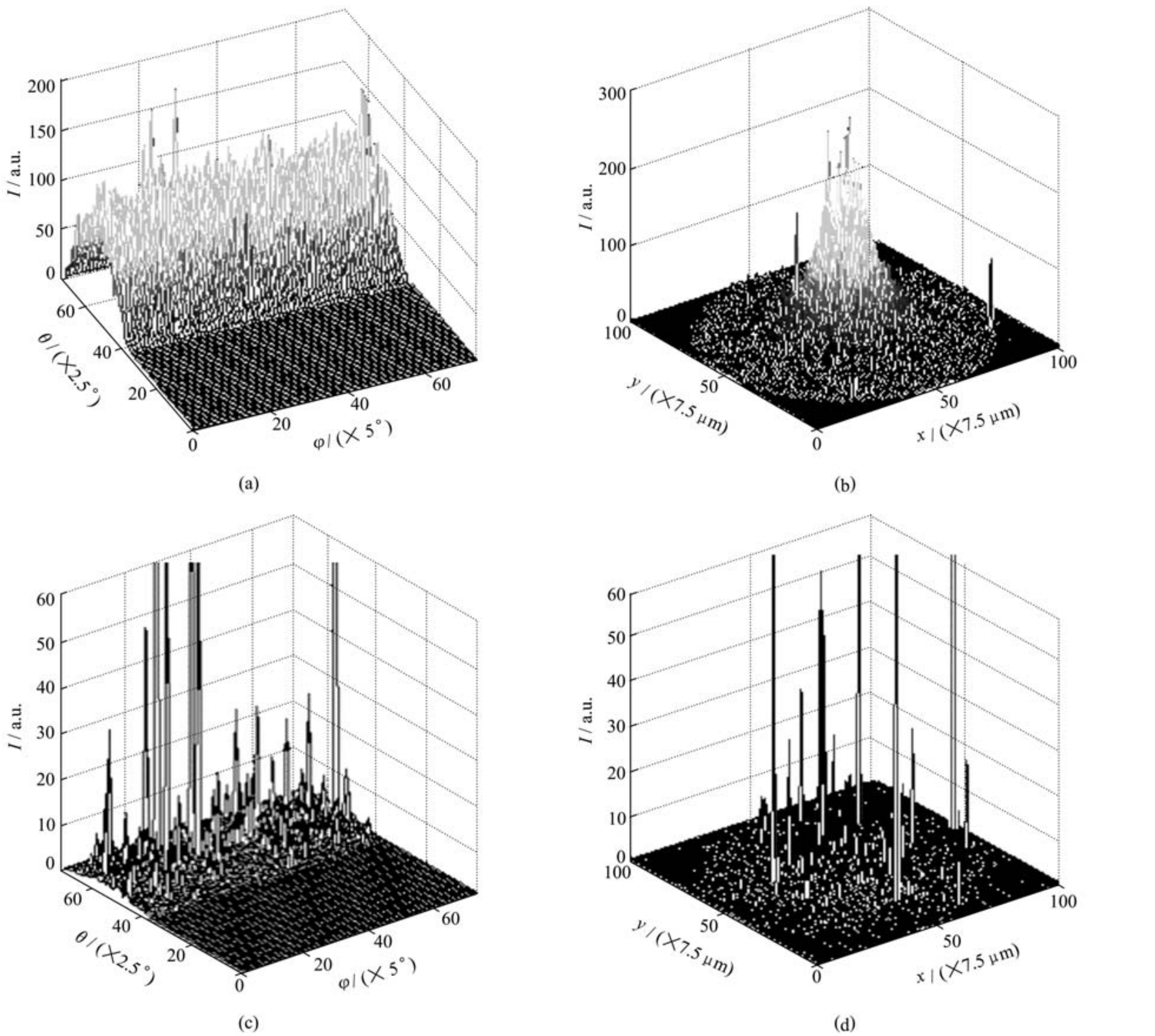

Fig. 2 When $E_{\text{inc}} = 0.5$ μJ, angle (a) and position (b) distributions of emission; when $E_{\text{inc}} = 2.3$ μJ, angle (c) and position (d) distributions of emission

media. At certain locations, the spectrum is not monochromatic and has a spectrum range, shown in Fig. 3. The spatial locations and directions of emission of a single frequency spread widely too, as shown in Fig. 4.

4.2 Discussion on distinct sharp peaks

In Fig. 5, the number distribution of photons and light intensity (product of weight and photon number) contributed from a different number of scattering events are plotted. Comparing Figs. 5 (a) and 5(b), it can be easily observed that the spectrum of nonresonant random laser becomes narrow markedly and it can also be concluded how gain spectrum and the number of scattering events influence the random laser spectrum. Low gain coefficients lead to a short resident time and small number of scattering events in the random media. Thus, the light at the side wavelength of the gain spectrum, which has low gain coefficient, is amplified slightly and contributes little to the intensity. On the contrary, photons around the peak gain wavelength with larger gain coefficients experience more scattering events and stay in the amplifying random media for a longer time, and then they are amplified more. Finally, the entire random laser spectrum narrows evidently and sharp intensity peaks appear. The wavelengths of those intensity peaks are around the peak wavelength of gain spectrum, as shown in Fig. 5(b). In addition, from Fig. 5 the origin of these intensity peaks can be easily found. Figure 5(b) shows that the maximal intensity peaks are contributed from those photons around the peak gain wavelength, which experience rather more scattering

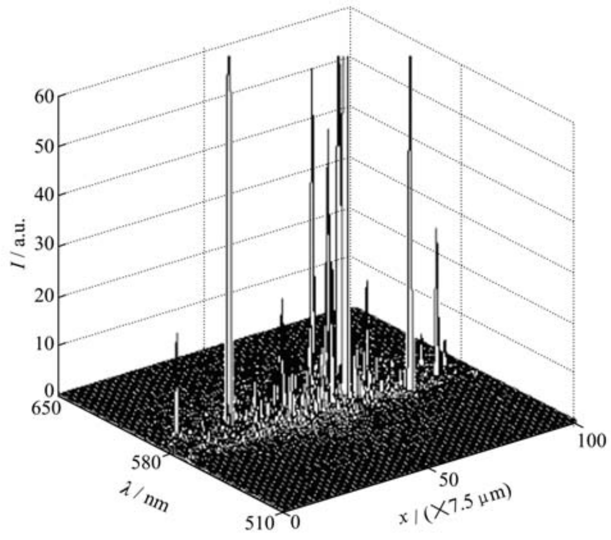

Fig. 3 Spectra at different spatial positions when $E_{inc} = 2.3$ μJ

events, but the number of these photons is very small as shown in Fig. 5(a). Therefore, rare photons experience many scattering events and take a longer path in amplifying random media, which produces distinct sharp intensity peaks on the top of the smooth spectrum background. When excitation energy is small, photons travel in random media with a small gain, and longer path scattering has a negligible contribution to the entire light intensity. When excitation energy increases, gain enhances accordingly, and photons taking a longer path in random media are amplified enough and have a dominant contribution to the entire intensity. In a word, the distinct sharp peaks of the entire emission spectrum are caused by the gain coefficient and number of scattering, which coincides with the result of Ref. [8].

In addition, from Fig. 1, another phenomenon can be found. Sharp peaks appear at the range of longer wavelength when excitation energy is small, and as excitation energy increases, sharp peaks shift to the shorter wavelength. The reason is that when the scatter size is constant, photons with different wavelength experience different scattering strength, and photons with shorter wavelength are scattered relatively more strongly and experience more scattering events. When excitation energy is small, the scattering numbers at all wavelengths are not large, and the gain coefficient has more influence on the determination of the position of the wavelength of sharp intensity peaks. On the other hand, when strong excitation energy is used, the scattering number dominates the sharp peak wavelength. Therefore, with strong excitation, sharp peaks appear at shorter wavelength where photons experience more scattering events.

5 Conclusions

In this article, Monte Carlo technique has been used to simulate the nonresonant emission of amplifying random media. The existence of scatterers makes random laser different from conventional ASE. On the other hand, the disorder of scatterer position and angle distribution also makes the random laser completely different from conventional laser. All these results are consistent with the obtained results by other researchers. In addition,

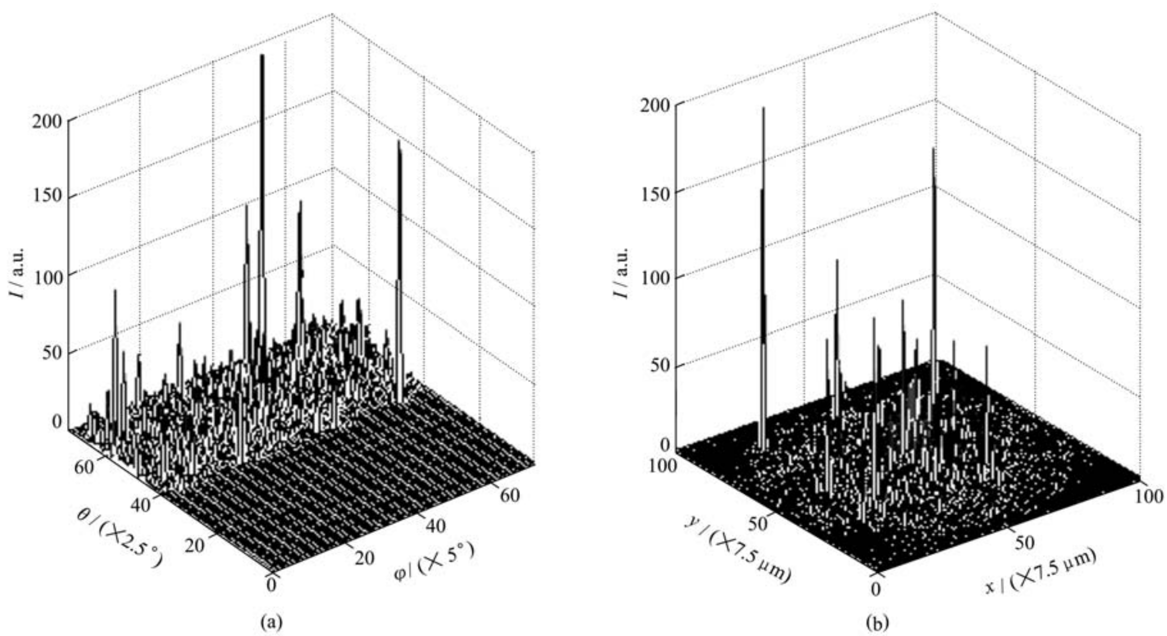

Fig. 4 Angle and position distributions of single-frequency radiation at 580 nm when $E_{inc} = 1.5$ μJ

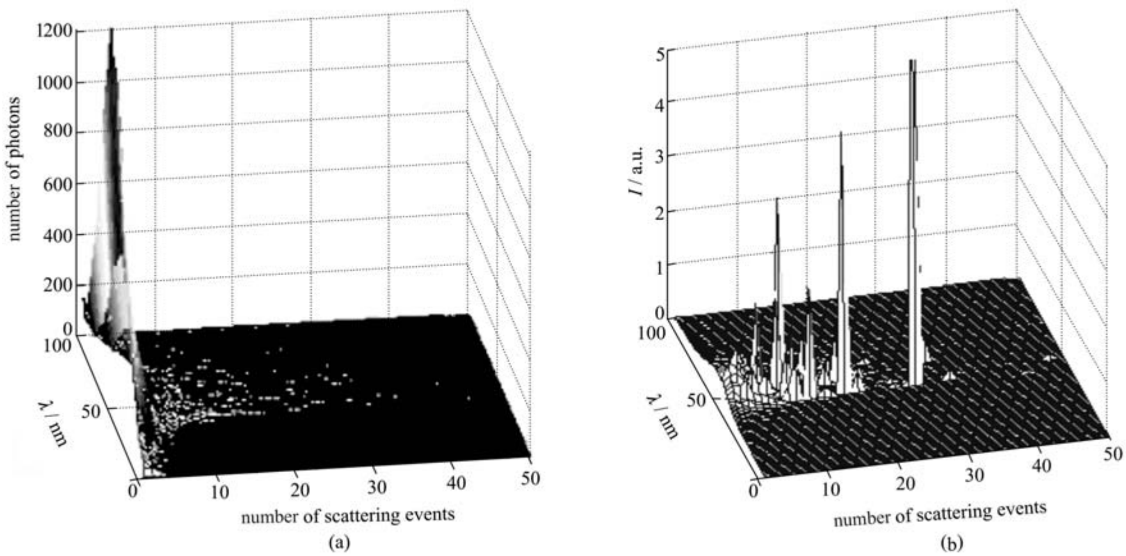

Fig. 5 Distribution of photons experiencing (a) and emissions contributing from (b) different numbers of scattering events when $E_{\text{inc}} = 3\ \mu\text{J}$

through calculation, it is found that the emission at a certain location in random media is not monochromatic and covers wide spectrum range, and that of a single frequency spread widely in direction and location. These results show the characteristics of random laser from another angle. The narrowing of random laser spectrum is caused by gain coefficient and number of scattering events. Rare photons experience rather more scattering events and take a longer path, which produces distinct sharp peaks on the smooth spectrum background. In this article, the simulation is only carried out in the condition of $\tau \ll \tau_0$, and the simulation is divided into two independent stages: pumping stage and emission stage. When this condition is not satisfied, the simulation could not be divided into two independent stages and they should be carried out by turns.

References

1. Letokhov V S. Stimulated emission of an ensemble of scattering particles with negative absorption. JETP Letters, 1967, 5(8): 212–215
2. Letokhov V S. Generation of light by a scattering medium with negative resonance absorption. Soviet Physics JETP, 1968, 26(4): 835–839
3. Noginov M A, Bahoura M, Noginova N, et al. Study of absorption and reflection in solid-state random laser media. Applied Optics, 2004, 43(21): 4237–4243
4. Lawandy N M, Balachandran R M, Gomes A S L, et al. Laser action in strongly scattering medium. Nature, 1994, 368(6470): 436–438
5. Cao H, Xu J Y, Zhang D Z, et al. Spatial confinement of laser light in active random media. Physical Review Letters, 2000, 84(24): 5584–5587
6. Zhang Y, Wang G, Cui Y P, et al. Electrochemical deposition and stimulated emission of zinc oxide thin films. Chinese Journal of Lasers, 2004, 31(1): 97–100 (in Chinese)
7. Cao H, Xu J Y, Chang S H, et al. Transition from amplified spontaneous emission to laser action in strongly scattering media. Physical Review E, 2000, 61(2): 1985–1989
8. Mujumdar S, Ricci M, Torre R, et al. Amplified extended modes in random lasers. Physical Review Letters, 2004, 93(5): 053903-1–053903-4
9. Wang C, Liu J S. Polarization dependence of lasing modes in two-dimensional random lasers. Physics Letters A, 2006, 353(2–3): 269–272
10. Mujumdar S, Cavalieri S, Wiersma D S. Temperature - tunable random lasing: numerical calculations and experiments. Journal of the Optical Society of America B, 2004, 21(1): 201–207
11. Schafer F P. Dye Lasers (in Chinese, trans. Chen Changmin, Sun Mengjia, Su Dachun). Beijing: Science Press, 1987, 92–174
12. Niginov M A, Bahoura M, Noginova N, et al. Stimulated emission in scattering and composite dielectric media (random lasers): effect of particle size. Proceedings of SPIE, 2003, 5218:124–139
13. van de Hulst H C. Light Scattering by Small Particles. New York: Jonh Wiley & Sons Press, 1957, 115–130
14. Cao H. Random lasers: development, features and applications. Optics & Photonics News, 2005, 16(1): 24–29