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Mode competition of bichromatic laser emission in two-dimensional strong scattering random medium

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ABSTRACT

The mode competition of the bichromatic laser emission in two-dimensional (2D) random medium was studied. Based on the time-dependent theory, a model for dual-wavelength random laser was established. The Maxwell equations and rate equations were combined and solved by using the finite different time domain (FDTD) method. Results show that the emission intensity of both wavelength ranges increases simultaneously with the increasing of the surface-filling fraction. The mode dominance of the bichromatic emission can be switched by changing the scatterer radius. The emission intensity of the longer wavelength increases when enlarging the excitation area. The controllability of these lasing modes may provide a potential application of optical switches.

1. Introduction

Since the prediction by Letokhov, random laser action has been widely studied for more than 40 years [1–11]. Different from a conventional laser, its cavity is not formed by some regular mirrors. Instead, the optical feedback comes from light scattering in disordered media, and the interference effect gives rise to resonant modes at specific frequencies. These modes are not regularly structured and equally spaced in frequency-domain. Experimental and theoretical studies show that the mode structure of random lasers rely on the sample parameters, including the sample size, shape, scatterer size, surface-filling fraction, an so on. Previous studies usually focus on the gain media which have single emission wavelength [4–9]. In recent experiments, a bichromatic emission phenomenon was found in the random laser system containing optically pumped Rhodamine 640 solutions and TiO_2 nanoparticles [12–15]. The spectral properties were investigated as a function of the dye concentration, scatterer density, and pumping area in these experiments. Results show that the emission intensity ratio of these two wavelength ranges was influenced mainly by the sample and pumping parameters.

In order to explain this dual-color laser emission phenomenon, John and Pang propose a model for dye system with excited singlet (S_0, S_1)and triplet (T_1, T_2) electronic energy levels [16]. They suggested that $S_1 \rightarrow S_0$ and $T_2 \rightarrow T_1$ represent the shorter and longer wavelength emission process, respectively. Later Balachandran and Lawandy consider that the reabsorption process of the primary lasing emission will result in the secondly lasing at longer wavelength [13]. Different form the previous viewpoint, Vaveliuk and his coworkers present a new model for bichromatic laser emission from laser dye solutions containing randomly distributed scattering particles [17]. They suggested that bichromatic emission is produced by two separate aggregate: monomers and dimers. With these models, most of the experimental results can be explained.

The key assumption of John's model is that there is a certain number of dye molecules in the triplet fundamental state, but this is wrong since the nanosecond pulse setup used in the experiments makes the triplet build-up effects negligible. On the other hand, Balachandran neglect the dimmer formation based on measurements of absorption and fluorescence spectra of pure dye solutions [17]. In Vaveliuk's theory, the dynamics of the dual-color laser emission was modeled by a set of rate equations, whose solutions were agree with the experimental data. However, this is not a time dependent theory and the temporal properties of the emission in the system cannot be described directly. In order to create a strict multi-wavelength random lasing model, the Maxwell equations must be used to describe the different light fields, which corresponding to the laser emissions at different wavelength. Based on the time dependent theory of the random laser and the "monomer-dimer" model presented by Vaveliuk, we have established a new theory model. This model creates a new energy system and simplifies the internal process of the dye molecules in the random media. In more recent years, Liu has propose an analogous model to study the dual-color laser emission in disordered media [18,19]. However,

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he has either considered the diffusion regime in 2D random media, or the mode competition controlled by local pumping for 1D case. In this paper, we have established a more accurate time dependent model, especially in the description of the reabsorption process. We consider the strongly scattering regime and focus on the mode competition of bichromatic laser emission in two-dimensional active random medium controlled by sample parameters. Results show that the density of scattering particles, scatterer radius and excitation area play an important role in the competition behavior.

2. Theoretical model

The two-dimensional (2D) square random system with size L^2 in the x–y plane is considered in our work. It consists of circular particles with a radius r and refractive index n_2, which are randomly distributed in an active medium with a refractive index n_1. This system can be considered as the multiple scattering light is confined in the x–y plane among a 3D sample, which resulting in a quasi-2D type of light transport. The density of scattering particles can be defined as the surface-filling fraction $\Phi = N\pi r^2/L^2$, where N is the amount of the particles.

According to Vaveliuk's theory, there exist two relative fluorescent aggregates in the random system, and the energy exchange between them includes absorption process and non-radiative energy transfer (NET) process from the monomers to dimers. The new energy level system used in our work is shown in Fig. 1. It contains five energy levels but actually two sets of four-level energy system, which corresponding to the monomer and the dimer respectively. These two lasing processes share a same energy level (level 5) and the energy exchange between these two sets of four-level energy system can be achieved via the monomer-dimer energy transfer process (level 5–level 4). Based on Vaveliuk's experimental results, the transition probability from level 5 to level 2 and 4 ($1/\tau_{52}$ and $1/\tau_{54}$) are proportional to the concentration of monomer and dimer, respectively. Therefore, the dye concentration of the system can be described by the coefficient $C = \tau_{52}/\tau_{54}$. The external light excite the electrons from level 0 to level 5 with the pumping rate W_{p1} for the monomer and from level 0 to level 4 with the pumping rate W_{p2} for the dimer. W_{p1} is a fixed value and W_{p2} can be written as $W_{p2} = K_1 W_{p1} + K_2 E_1$, while $K_1 W_{p1}$ is the external pumping rate and $K_2 E_1$ represent the reabsorption process of the dimer. Since the absorbed light comes from the monomer, the corresponding part $K_2 E_1$ is proportional to the electrical-field intensity E_1 of the emission light. The rate equations can be expressed as follows:

$$\frac{dN_5}{dt} = W_{p1} \cdot N_0 - \frac{N_5}{\tau_{52}} - \frac{N_5}{\tau_{54}} \tag{1}$$

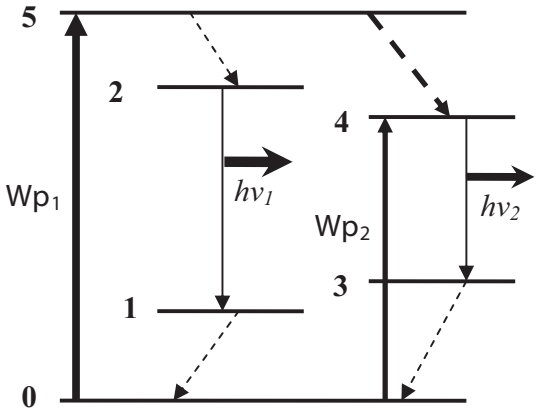

Fig. 1. Scheme of the bichromatic laser energy level system.

$$\frac{dN_4}{dt} = \frac{N_5}{\tau_{54}} - \frac{N_4}{\tau_{43}} + \frac{E_2}{h\nu_2} \cdot \frac{dP_2}{dt} + W_{p2} \cdot N_0 \tag{2}$$

$$\frac{dN_3}{dt} = \frac{N_4}{\tau_{43}} - \frac{N_3}{\tau_{30}} - \frac{E_2}{h\nu_2} \cdot \frac{dP_2}{dt} \tag{3}$$

$$\frac{dN_2}{dt} = \frac{N_5}{\tau_{52}} - \frac{N_2}{\tau_{21}} + \frac{E_1}{h\nu_1} \cdot \frac{dP_1}{dt} \tag{4}$$

$$\frac{dN_1}{dt} = \frac{N_2}{\tau_{21}} - \frac{N_1}{\tau_{10}} - \frac{E_1}{h\nu_1} \cdot \frac{dP_1}{dt} \tag{5}$$

$$\frac{dN_0}{dt} = \frac{N_1}{\tau_{10}} + \frac{N_3}{\tau_{30}} - W_{p1} \cdot N_0 - W_{p2} \cdot N_0 \tag{6}$$

where E_1 and E_2 are the electrical-field intensity of the shorter and longer wavelength emission, respectively. P_1 and P_2 are the corresponding electric polarization density. The center frequency of these two lasing emissions are chosen as $\nu_1 = 4.84 \times 10^{14}$ Hz ($\lambda_1 = 620$ nm) and $\nu_2 = 4.62 \times 10^{14}$ Hz ($\lambda_1 = 650$ nm).

We considered a 2D transverse magnetic (TM) field in the x–y plane, thus the Maxwell's equations are described as before [9]. The polarization equations read

$$\frac{d^2 P_{iz}}{dt^2} + 2\pi \cdot \Delta\nu_i \frac{dP_{iz}}{dt} + 2\pi \cdot \nu_i^2 P_{iz} = \kappa_i \Delta N_i E_{iz} \quad (i = 1, 2) \tag{7}$$

where $\Delta N_1 = N_1 - N_2$ and $\Delta N_2 = N_3 - N_4$ are the population density difference between the lower and upper levels of the atomic transition, $\Delta\nu_1 = 2\pi(1/\tau_{21} + 2/T_2)$ and $\Delta\nu_2 = 2\pi(1/\tau_{43} + 2/T_2)$ are the linewidth where T_2 is the collision time. The constants $\kappa_1 = 3\varepsilon_0 c^3/(2\pi\nu_1^2\tau_{21})$ and $\kappa_2 = 3\varepsilon_0 c^3/(2\pi\nu_2^2\tau_{43})$ are relate to the energy levels. The values of the other parameters used in the above equations are chosen as: $\tau_{21} = \tau_{43} = 1 \times 10^{-9}$ S, $\tau_{10} = \tau_{30} = 10^{-11}$ S, $\tau_{52} = \tau_{54} = 10^{-13}$ S, $T_2 = 2 \times 10^{-14}$ S. By use of the finite-difference time-domain (FDTD) method and the PML (perfectly matched layer) absorbing conditions, the electromagnetic fields can be calculated. The Fourier transformation is carried out to obtain the power spectrum. The space and time increment are chosen to be $\Delta x = \Delta y = 10$ nm and $\Delta t = \Delta x/(2c) \approx 1.67 \times 10^{-17}$ S.

3. Results and discussion

In what follows, we will analyze the mode competition of the bichromatic laser emission under different system parameters, including the surface-filling fraction, scatterer size and excitation area.

3.1. Surface-filling fraction

In a 2D random system, the density of the scatterers is described by surface-filling fraction Φ. This parameter plays a key role in the light localization: once the surface-filling fraction exceed a critical value, the transition from diffusion state to localization state occurs. In this subsection, we select a 2D sample with $S = 5 \times 5\ \mu m^2$, $r = 60$ nm, $n_1 = 1.4$, $n_2 = 2.7$ and $\tau_{52} = \tau_{54} = 10^{-13}$ S. The pumping parameters are selected as $W_{p1} = 1 \times 10^{13}\ s^{-1}$ and $K_1 = K_2 = 0.5$. Fig. 2 shows the spectral intensity of the bichromatic laser emission when changing the surface-filling fraction. As can be seen, the spectral intensity of the both wavelength ranges (620 nm and 650 nm) are weak at a small surface-filling fraction ($\Phi = 30\%$). With the increase of the surface-filling fraction, the spectral intensity of both ranges increase and the number of the peaks decrease simultaneously. The reason is that with a larger surface-filling fraction, the multiple scattering process becomes more intensive, which will increase the light localization in certain area. With the interplay between localization and amplification, some lasing modes are amplified around the respective central frequencies. Due to the mode competition, only a limited number of lasing modes can exist in the case of large amplification.