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Remarkable spectral difference between different polarized modes in four-level two-electron atomic system

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A R T I C L E I N F O

ABSTRACT

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1. Introduction

The theoretical and experimental work on the propagation of the light waves in random systems has been studied widely [1-24]. The general spectral signatures of random lasing are an overall narrowing of the emission spectrum, a threshold behavior, and the eventual presence of narrow spikes in the emission spectrum. The distinctive properties obtained from early theoretical and experimental efforts generated great enthusiasm to hunt for additional examples of random lasers and, indeed, many have emerged in different material systems [2–10]. Typical random laser systems usually select the dye as gain material and semiconductor material as scattering particles [5,6]. In these systems, optical feedback is provided by scattering of light due to the spatial inhomogeneity of the refractive index. Several models have been set up in the theoretical study of the lasing process in active random media [11–15]. Especially, a typical one-dimensional (1D) FDTD model was built by Jiang and Soukoulis [12] and was extended to 2D TM case by Sebbah and Vanneste [13]. By the use of this model, much random lasing property has been explored in many investigations [15–17]. In 2002, Ito and Tomita performed an experiment in a 2D amplifying and scattering medium consisted of random array of dye-doped plastic fiber [18] and demonstrated that the random lasing radiation is polarization-dependent.

In some special random laser systems, the semiconductor materials such as ZnO or TiO_2 powder or film not only as the scattering materials, but also provide optical gain. In this case, the gain

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0030-4026/\$ - see front matter © 2013 Elsevier GmbH. All rights reserved. http://dx.doi.org/10.1016/j.ijleo.2013.06.051 behavior is quite complicated because the energy band structure of the semiconductor materials is different from the ordinary single atomic system [2,3,16]. The general models for the random laser, in which some semiconductor properties are neglected, cannot be used to study the temporal dynamics of the electric field exactly. The conduction and valence bands of the semiconductor materials are continuous sets of energy levels. An electron in the valence band can jump to the conduction band by absorbing a pump photon. This electron quickly loses energy and decays to the lowest energy level of the conduction band. Subsequently, the electron can decay to the top of the valence band by emitting a photon. Finally, the electron can decay to the unoccupied lower energy level of the valence band. Based on the semiconductor band structure, an effective FDTD semiconductor model is established to reveal the physical characteristics of coherent feedback in semiconductor random laser. In 2004, Shih-Hui Chang and Aleen Taflove presented a computational model of electrons dynamics and gain in four-level two-electron atomic system [16]. Based on the theory, the adoption of the two-electron model provides us with an opportunity: to use the two-electron atomic systems to study the spectral difference between different polarized light waves from 2D random media. The new model incorporates a semiconductor band structure which allows four energy levels for each of the two interacting electrons. Transitions between the energy levels are governed by coupling rate equations and the Pauli Exclusion Principle.

The spectral difference between different polarized modes in two-dimensional (2D) four-level two-

electron atomic system is investigated by simultaneously solving Maxwell's equations and rate equations

of electronic population. A new physical model including pumping dynamics is introduced, and the transitions between the energy levels are governed by coupling rate equations and the Pauli Exclusion Principle. Results indicated that polarized lasing modes have different field pattern, spectral range, and intensity.

The photons in TE field are easier to leak from random media than those in TM field. The quality factor of

TM modes is larger than that of TE modes and the loss of TM modes to be lower than that of TE modes.

The spectral properties in four-level two-electron atomic system are polarization-dependent.

By use of the updated model, the polarization-dependent lasing action in the semiconductor media can be investigated for both TE form (TE fields mean the electric and magnetic components to be within and perpendicular to the 2D plane) and TM form (TM fields mean the electric and magnetic components of the light wave to be perpendicular to and within the 2D plane). In order to investigate











Fig. 1. The sketch map of the structure of random medium.

the spectral difference between different polarized modes, first things first, it is necessary to research power spectrum of random media and the spectral time evolution under different pumping rate. A general model is created to analyze the characteristics of both TE and TM polarized state respectively. With this semi classical approach, we treat the atom quantum mechanically, and the electromagnetic wave classically. This model could be potentially further extended to analyze other random lasing phenomenon.

2. Theoretical model

The 2D disordered sample is described as shown in Fig. 1. It is the simplification of real experiments including a random collection of cylinders oriented along the *z* axis. When the system parameters (i.e.,*l*,*r*, ε_1 , ε_2) are given, they can create a number of random media. The proposed model is based on the incorporation, in Maxwell's equations, of the coupled rate equations with Pauli Exclusion Principle, taking into account the dynamic pumping in the FDTD formulation. This model permits investigation of both the transient processes of semiconductor random lasers as well as their optical behaviors at longer time scales. Therefore, we use the FDTD method to obtain the spectrum of each polarized mode in 2D random media.

In isotropy, non-magnetic disordered media, Maxwell's equations read as:

$$\nabla \times \vec{\mathbf{E}} = -\mu_0 \frac{\partial \dot{\mathbf{H}}}{\partial t},\tag{1.1}$$

$$\nabla \times \vec{\mathbf{H}} = \varepsilon_0 \varepsilon_r \frac{\partial \vec{\mathbf{E}}}{\partial t} + \frac{\partial \vec{\mathbf{P}}}{\partial t}, \qquad (1.2)$$

where \mathbf{P} is the electric polarization density from which the amplification or gain can be obtained; ε_0 and μ_0 are the electric permittivity and the magnetic permeability of vacuum respectively, and ε_r is the relative dielectric constant. To account for the PEP (Pauli Exclusion Principle), a two-electron model is adopted in the four-level system. When the electron population in level *i* is close to one, electrons from the other levels are less likely to decay to level *i* because of the exclusion principle. Fig. 2 illustrates the transition process in a simplified four-level two-electron model.

Here, Levels 0 and 1 are in the valance band and Levels 2 and 3 are in the conduction band. These four levels are treated as two coupled dipole oscillators. Levels 0 and 3 correspond to dipole $\vec{P}_{\rm b}$, and Levels 1 and 2 correspond to dipole $\vec{P}_{\rm a}$. To simplify the analysis, the $\vec{E}/\hbar\omega_{\rm b} \times d\vec{P}_{\rm b}/dt$ is replaced by $W_p \cdot N_0$ to describe the optical pump, W_p is the pump rate which can be seen from Fig. 2. The following are the rate equations for the electron populations in the four levels:

$$\frac{dN_3}{dt} = -\frac{N_3(1 - N_2/N_2^0)}{\tau_{32}} - \frac{N_3(1 - N_0/N_0^0)}{\tau_{30}} + W_p \cdot N_0$$
(2.1)



Fig. 2. The sketch map of Four-level two-electron model.

$$\frac{dN_2}{dt} = \frac{N_3(1 - N_2/N_2^0)}{\tau_{32}} - \frac{N_2(1 - N_1/N_1^0)}{\tau_{21}} + \frac{\tilde{E}}{\hbar\omega_a} \cdot \frac{d\tilde{P}_a}{dt}$$
(2.2)

$$\frac{dN_1}{dt} = \frac{N_2(1 - N_1/N_1^0)}{\tau_{21}} - \frac{N_1(1 - N_0/N_0^0)}{\tau_{10}} - \frac{\vec{E}}{\hbar\omega_a} \cdot \frac{d\vec{P}_a}{dt}$$
(2.3)

$$N_0 = N_{\text{total}} - N_1 - N_2 - N_3 \tag{2.4}$$

where $\vec{E} = E_x \mathbf{e}_x + E_y \mathbf{e}_y + E_z \mathbf{e}_z$, $\vec{P}_a = P_x \mathbf{e}_x + P_y \mathbf{e}_y + P_z \mathbf{e}_z$, with \mathbf{e}_x , \mathbf{e}_y , \mathbf{e}_z being the unit vectors in the *x*, *y* and *z* directions, respectively. In these equations, $v_a = \omega_a/2\pi = 6.45 \times 10^{14}$ Hz ($\lambda_a = 464.46$ nm), N_i is the electron population density in level *i*, *i* = 0 to 3, τ_{ij} is the lifetime of levels *i* and *j*. The electron populations vary with pumping $\vec{E} \cdot d\vec{P}_a/dt$ and the spontaneous emission decay $(N_i - N_j)/\tau_{ij}$. The spontaneous decay rate is reduced by a factor $1 - N_i/N_i^0$ based on PEP, where N_i/N_i^0 is the normalized density of the population in the lower energy level. The polarization equation should be described as follows:

$$\frac{d^2 \vec{P}_a}{dt^2} + \Delta \omega_a \frac{d \vec{P}_a}{dt} + \omega_a^2 \vec{P}_a = \kappa_a (N_2 - N_1) \vec{E}$$
(3)

where \vec{P}_a has the resonant frequencies ω_a ; $\hbar\omega_a$ is the en difference between Levels 1 and 2. There-into, $\kappa_a = 6\pi\varepsilon_0 c^3/(\omega_a^2\tau_{21})$, $\Delta\omega_a = 1/\tau_{21} + 2/T_2$; For TM state, $\vec{P} = P_z \mathbf{e}_z$ and $\vec{E} = E_z \mathbf{e}_z$, while for TE state, $\vec{P} = P_x \mathbf{e}_x + P_y \mathbf{e}_y$ and $\vec{E} = E_x \mathbf{e}_x + E_y \mathbf{e}_y$. The values of those parameters in the equations that will be used in simulating the active part in the following numerical calculations are taken as: $T_2 = 2 \times 10^{-14} s$, $\tau_{21} = \tau_{30} = 3 \times 10^{-10} s$, $\tau_{10} = \tau_{32} = 10^{-13} s$, the initial population density is $N_i^0 = 5 \times 10^{23} m^{-3}$.

3. Results and discussions

In order to exhibit the spectral difference between different polarized modes, we select a 2D random medium with l = 5.5 µm, Φ = 40%, r = 60 nm, n_1 = 1, and n_2 = 2 to calculate the spectral intensity varying with the wavelength at different pump rates. For convenience, we were assuming that all semiconductor particles in the 2D *x*-*y* plane axis. The calculated power spectra at t = 8 ps for the two polarization states under different pump rates are shown in Fig. 3.

It revealed clearly that the spectral intensity of each polarization state growing up as the increasing of pump rate. With the increasing pumping intensity, there are more electrons were pumped to the upper energy level per unit time. The population inversion would be achieved more easily. So the lasing modes will appear earlier, along with a narrower and more intense lasing pulse. We choose the mode that has minimum lasing threshold. The wavelength of marked TM mode is λ_{TM} = 464.71nm and the wavelength of marked TE mode is λ_{TE} = 464.54nm. It is easily understandable because the central wavelengths of marked modes are very near the transition wavelength λ_a (464.46 nm) of the random medium. If the pump is strong enough, many other polarized modes would be stimulated.

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