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Numerical study on upconversion random lasing in strong scattering disordered medium

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ABSTRACT

The upconversion random lasing properties in two-dimensional (2D) disordered medium was studied. Based on the time-dependent theory, a model for upconversion 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 at 381 nm is determined by the pumping intensity. For low pumping rates, the 381 nm signal is mainly contributed from the transition $^4D_{3/2} \rightarrow {}^4I_{11/2}$ while the transition $^4D_{7/2} \rightarrow {}^4I_{13/2}$ is negligible. When the pumping rate increases to a certain value, the intensity of these two upconversion signals are comparable. The duration of the pulse from the transition $^4D_{7/2} \rightarrow {}^4I_{13/2}$ is short while the emission from $^4D_{3/2} \rightarrow {}^4I_{11/2}$ has a long fall time. As the pumping rate increases, the width and the peak interval of two pulses will decrease simultaneously. The simulation results may provide a control method of upconversion random lasers.

1. Introduction

Random laser was predicted by Letokhov in 1968 and receiving large attention for its unique physical mechanism and potential applications in recent years [1–4]. The cavity of a random laser is not formed by regular mirrors. Instead, the optical feedback comes from multiple light scattering in disordered media, while the interference effect gives rise to resonant modes at particular frequencies. The random lasing properties are quite different from those of a conventional laser. Lasing from strongly scattering media has been realized in various systems such as nanoparticles of laser crystals [5], dye solutions containing small particles [6], polymers [7,8] and semiconductor powders [9,10]. The mode structure of a random laser is not regular and the output beam is not unidirectional. However, these unique characteristics lead to many promising applications, such as laser displaying, sensing, integrated light source, and so on. The random lasers mentioned above are based on one-photon pumping and emission wavelengths are in the Stokes side of the excitation pulse, which means that the random lasing wavelength is longer than the pumping one. In order to provide different emission wavelengths to meet these various demands, researches have exploited upconversion lasing technique by using multiphoton excitation process in different random laser materials [11–13]. The advantage of the upconversion random laser is that the emitted light can cover the ultraviolet (UV) regime by using an excitation laser operating in visible or near-infrared (NIR) range. This will lead to some potential applications such as full-filed imaging and cancer detection.

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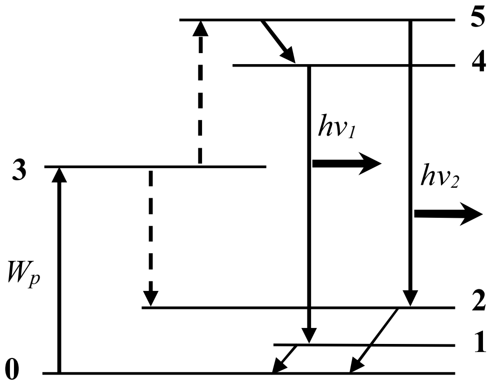

Fig. 1. Scheme of the upconversion laser system based on cross-relaxation.

In recent years, upconversion random lasers based on Nd^{3+} doped fluoroindate glass [14] or nanocrystalline powder [15] were demonstrated, a violet or red emission was obtained respectively. In order to explore the deeper physical mechanism of this phenomenon, researchers have proposed a simple model to describe the kinetics of the upconversion process. They suggested that the upconversion emission is attributed to energy transfer between coupled excited Nd^{3+} ions, i.e. cross-relaxation process. The Nd^{3+} ions were resonantly excited to level $^2G_{7/2}$, then the pairs of coupled ions will exchange energy in the way that one ion is promoted to level $^4D_{7/2}$ while the other decays to level $^4I_{13/2}$. There are two pathways for the ions at level $^4D_{7/2}$ to decay and emit at 381 nm, one is directly from level $^4D_{7/2}$ to $^4I_{13/2}$, the other is from $^4D_{7/2}$ to $^4D_{3/2}$ by nonradiative decay and then to level $^4I_{11/2}$. By neglecting the mixing of individual ions' states, Oliveira has written some equations to describe the dynamics of the anti-Stokes process among the pair states. However, this simple model can only be used to investigated the population density evolution of each energy level and the temporal behavior of the upconversion signal. Random laser is a special class that the multiple scattering process plays an important role in the lasing process, which determine the lasing modes. The feedback mechanism was provided by the multiple light scattering and gain mechanism is obtained by external pumping. A complete model for random laser should include the dynamics of gain and feedback mechanism. By combining the Maxwell equations and rate equations, researchers have developed a time dependent theory for random lasers [16,17]. This model has successfully explained many characteristics of random lasing, including the spatial distribution, emission spectrum, lasing threshold, temporal evolution, and so on. In a strict upconversion random lasing model, one must couple the Maxwell equations with the rate equations to describe the light fields, which corresponding to different decay process. Based on the time dependent theory and the cross-relaxation scheme, we have built a new theory model. This model creates a new energy system to describe the cross-relaxation process of the coupled ions. In this paper, we consider the strongly scattering regime for Nd^{3+} doped active random medium. By changing the pumping and sample parameters, the spectral, spatial and temporal characteristics were studied.

2. Theoretical model

We consider a two-dimensional (2D) square random media with size L^2 in the x–y plane. There are N circular particles with radius r and refractive index n_2 randomly distributed in the medium with an refractive index n_1. The light is confined in this plane and result in a quasi-2D light transport. The density of scattering particles can be defined as the surface-filling fraction $\Phi = N\pi r^2/L^2$, which will affect the strength of multiple light scattering.

Based on the simplified energy level scheme of Nd^{3+} ions and corresponding upconversion process in pair states present by Oliveira, a new energy level system was created in our work and shown in Fig. 1. It contains six energy levels but actually two coupled energy systems for pair ions. The pathway from ground state to state $^4D_{7/2}$ by one-photon absorption is discarded, therefore we must consider the two-step process. The electrons in the ground state (level 0, $^4I_{9/2}$) are transferred to an intermediate state (level 3, $^2G_{7/2}$) by an external pump at a fixed rate Wp. The cross-relaxation process between two coupled ions will exchange energy in such way that one ion is excited to state $^4D_{7/2}$ while the other decays to state $^4I_{13/2}$. As it happens in a pair of ions, the amount of electrons from level 3 to level 5 ($^4D_{7/2}$) or level 2 ($^4I_{13/2}$) is equivalent if neglecting the energy loss. This process is denoted in Fig. 1 with two dashed arrows. Electrons in level 5 flow downward to level 4 ($^4D_{3/2}$) by means of nonradiative decay process with a characteristic time τ_{54}. The laser transition takes place from level 4 to level 1 ($^4I_{11/2}$) with an emission frequency ν_1. There is another pathway that laser action take place directly from level 4 to level 2, with a frequency ν_2. The electrons in levels 1 and 2 will decay rapidly into the ground state (level 0, $^4I_{9/2}$) with the characteristic time τ_{10} and τ_{20}, respectively. Although the emission frequency of these two lasing processes is identical,