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2015 J. Phys.: Conf. Ser. 574 012077

(http://iopscience.iop.org/1742-6596/574/1/012077)

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Abstract. In this work, we show the resolution of the rate equations in powder random lasers by using the Crank-Nicholson finite difference method. Light propagation in our powders is described by the model of light diffusion. The generalized time-dependent random laser equations describing our system are formed by three differential coupled equations: two diffusion equations for the pump and emitted light and a rate equation for the density of the dopant molecules in the excited state. The system has been solved for two pumping schemes (one-photon and two-photon excitation) and for a wide range of temporal incident pulses (from femtoseconds to nanoseconds).

1. Introduction

Conventional lasers are usually constructed from two basic components: a gain material that is pumped in order to provide amplification of light and a cavity to provide feedback. However, random lasers replace the traditional laser cavity with a random, multiple-scattering medium. This type of laser becomes a subject of intense theoretical and experimental studies because of its important potential applications [1]. In addition, a major advantage of random lasers over regular lasers is that their production is cheap and the required technology relatively simple, but the laser dynamics are much more complex than that of the conventional laser and there is still much to understand. For example, scientists recently explored the mode-locking of random lasers and investigated how to control their operation.

The random laser equations have been solved by using several numerical methods due to the difficulty of finding the exact solution: method of lines [2], Montecarlo simulation [3] and finite-difference time domain method (FDTD) [4]. In this work, we solve these equations for two pumping schemes, one-photon (OP) and two-photon (TP) excitations, and for a wide range of temporal incident pulses (from femtoseconds to nanoseconds) by using the Crank-Nicholson finite difference method.

2. Theoretical model

Assuming a diffusive propagation of light in powder random lasers, the equations describing our model for one and two-photon excitations are:

$$\frac{\partial W_p(z,t)}{\partial t} = D_p \frac{\partial^2 W_p(z,t)}{\partial z^2} - g(z,t) + p(z,t) \tag{1}$$

$$\frac{\partial W_e(z,t)}{\partial t} = D_e \frac{\partial^2 W_e(z,t)}{\partial z^2} + f \, \mathrm{v} \, \sigma_{\mathrm{em}} N(z,t) W_e(z,t) + \gamma \frac{N(z,t)}{\tau_s} \tag{2}$$

$$\frac{\partial N(z,t)}{\partial t} = g(z,t) - f \, \mathrm{v} \, \sigma_{\mathrm{em}} N(z,t) W_e(z,t) - \frac{N(z,t)}{\tau_s} \tag{3}$$

where $W_p(z,t)$ and $\partial W_e(z,t)$ are the light densities at the pump and emission wavelengths respectively and $N(z,t)$ is the density of dye molecules in the excited state. The term corresponding to light absorption, *g(z,t)* is given by $g_{OP}(z,t) = f \, \mathrm{v} \, K_{abs} W_p(z,t)$ and by $g_{TP}(z,t) = f \, \mathrm{v}^2 \beta \hbar \omega_p W_p^2(z,t)$, for one and two- photon excitations respectively. K_{abs} is the one-photon absorption coefficient of the material at the pump wavelength and β is the two-photon one. The volume fraction, *f*, occupied by the scatters has been included in the equations to take into account the effective part of light density which penetrates into the particles. v is the speed of light in the medium. σ_{em} is the stimulated emission cross section, τ_s is the excited state lifetime and , D_p and D_e, are light diffusion coefficient for pump and emitted radiation, respectively. γ is the fraction of spontaneous emission contributing to the laser process. In both pumping schemes, the source of diffuse radiation, *p(z,t)*, is an incoming Gaussian pulse in the *z* direction which is extinguished along its path through the sample. This function *p(z,t)* is different for each type of excitation. The system of equations is solved with the following boundary and initial conditions [5]:

$$W_p(-l_e,t) = W_p(L+l_e,t) = W_e(-l_e,t) = W_e(L+l_e,t) = 0 \qquad \forall t$$

$$W_p(z,0) = \; W_e(z,0) = N(z,0) = 0 \qquad \forall z \tag{4}$$

l_e is the extrapolation length and L is the scattering sample thickness.

3. Numerical solution

The set of coupled non linear partial differential equations (1)-(3) are numerically solved by using the Crank-Nicholson method. On the condition that $n = \left(L + 2l_e\right)/h$ is an integer, the domain (*z, t*) is discretized by two sizes of step h (spatial) and *k* (time): $z_i = -l_e + ih \;\; 0 \le i \le n$ and $t_j = jk \;\; j \ge 0$.

3.1. One- photon excitation

The equation (1) is solved first and the derivatives are approximated at the mesh point by

$$\frac{\partial W_p(z_i,t_j)}{\partial t} \approx \frac{W_{i,j+1}^p - W_{i,j}^p}{k} \quad \text{with} \quad W_{i,j}^p = W_p(z_i,t_j)$$

$$\frac{\partial^2 W_p(z_i,t_j)}{\partial z^2} \approx \frac{1}{2}\left(\frac{W_{i+1,j}^p - 2W_{i,j}^p + W_{i-1,j}^p}{h^2} + \frac{W_{i+1,j+1}^p - 2W_{i,j+1}^p + W_{i-1,j+1}^p}{h^2} \right) \tag{5}$$

Replacing the derivatives (6) in the equation (1), evaluating $g(z,t)$ and $p(z,t)$ in the intermediate step between j and j +1 and using the boundary and initial condition, this equation can be written in matrix form as:

$$A^{OP}W_{j+1}^{p^*} = B^{OP}W_j^{p^*} + P_j^* \tag{6}$$

with $W_j^{p^*} \equiv \left(W_{1,j}^p, W_{2,j}^p, ..., W_{n-1,j}^p\right)^T$, $P_j^* = \left(p\left(z_1, t_{j+\frac{k}{2}}\right), p\left(z_2, t_{j+\frac{k}{2}}\right), ..., p\left(z_{n-1}, t_{j+\frac{k}{2}}\right)\right)^T$

where $A^{OP} = \left(a_{ij}^{OP}\right)_{1\le i\le n-1}$ and $B^{OP} = \left(b_{ij}^{OP}\right)_{1\le i\le n-1}$ are tridiagonal matrices

$$a_{ij}^{OP} = \frac{1}{k} + \frac{D_p}{h^2} + \frac{f\,\mathrm{v}\,K_{abs}}{2} \quad \text{if } i=j, \qquad a_{ij}^{OP} = -\frac{D_p}{2h^2} \text{ if } |i-j|=1, \qquad a_{ij}^{OP} = 0 \text{ if } |i-j|>1$$

$$b_{ij}^{OP} = \frac{1}{k} - \frac{D_p}{h^2} - \frac{f\,\mathrm{v}\,K_{abs}}{2} \quad \text{if } i=j, \qquad b_{ij}^{OP} = \frac{D_p}{2h^2} \text{ if } |i-j|=1, \qquad b_{ij}^{OP} = 0 \text{ if } |i-j|>1$$

Therefore, after solving the matrix equation (6), the solutions of equations (2) and (3) can be determined by the solution of the following matrix system:

$$\begin{cases} A_1^{OP} W_{j+1}^{e^*} = B_1^{OP} W_j^{e^*} + f\,\mathrm{v}\,\sigma_{em} N_j^{e^*} * W_j^{e^*} + \dfrac{\gamma}{\tau_s} N_j^{e^*} \\ N_{j+1}^{e^*} = N_j^{e^*} + k\left(f\,\mathrm{v}\,K_{abs} W_j^{p^*} - f\,\mathrm{v}\,\sigma_{em} N_j^{e^*} * W_j^{e^*} - \dfrac{1}{\tau_s} N_j^{e^*}\right) \end{cases} \tag{7}$$

where A_1^{OP} eta B_1^{OP} are the corresponding tridiagonal matrices and $N_j^{e^*} * W_j^{e^*}$ represents the element by element product of $N_j^{e^*}$ and $W_j^{e^*}$. In this case the functions of the coupled equations (2) and (3) are computed at the time gridpoints instead of at midpoints of the temporal subinterval in order to obtain a system of linear equations.

3.2. Two-photon excitation

The same process can be applied for two-photon excitations. Then, the equations (1)-(3) can be written as:

$$A^{TP}W_{j+1}^{p^*} = B_1^{TP}W_j^{p^*} - f\,\mathrm{v}^2\beta\hbar\omega_p W_j^{p^*} * W_j^{p^*} + P_j^* \tag{8}$$

$$\begin{cases} A_1^{TP} W_{j+1}^{e^*} = B_1 W_j^{e^*} + f\,\mathrm{v}\,\sigma_{em}\, N_j^{e^*} * W_j^{e^*} + \dfrac{\gamma}{\tau_s} N_j^{e^*} \\ N_{j+1}^{e^*} = N_j^{e^*} + k\left(f\,\mathrm{v}^2\beta\hbar\omega_p W_j^{p^*} * W_j^{p^*} - f\,\mathrm{v}\,\sigma_{em} N_j^{e^*} * W_j^{e^*} - \dfrac{1}{\tau_s} N_j^{e^*}\right) \end{cases} \tag{9}$$

4. Numerical results

We have carried out simulations to study the laser-like emission in several powder random lasers for OP [5, 6] and TP [7] excitations. The theoretical results are in a good agreement with the experimental data which validates the Crank-Nicholson method. As an example, we show the theoretical pulse shortening calculated from the reduction of the full width at half maximum (FWHM) of the temporal profiles when increasing the pump energies under TP excitation in a ground powder of a silica gel containing Rhodamine 6G doped silica nanoparticles. As the emitted photons are collected along the backward direction of the incident pump beam, the time evolution of the emitted light has been calculated from $\vec{F}_e(t) = -D_e \dfrac{\partial W_e(z,t)}{\partial z}\hat{z}$ evaluated at the front sample surface(z = 0). Figure 1 shows the FWHM of the emitted pulse obtained as a function of the pump pulse energy for different temporal

incident pulses. Δ represents the temporal half width at half maximum of incident pulse. The data have been fitted to a sigmoid function. The inflection points of these fits represent the threshold energy densities at which the light amplification begins. The dependence of the lasing threshold on the Δ incident pulse has been plotted in Figure 2. As it can be observed, lasing threshold increases very abruptly as the value of Δ is getting closer to the spontaneous lifetime of the material. The input values for these calculations are the material parameters: K_{abs} = 148.5 cm^{-1}, σ_{em} = 2.5x10^{-16} cm^2, τ_s (OP) = 1.65 ns, τ_s (TP) = 2.1 ns, $\beta\hbar\omega_p$ = 3.86x10^{-13} μm·ps, n_{eff} = 1.16, f = 0.43.

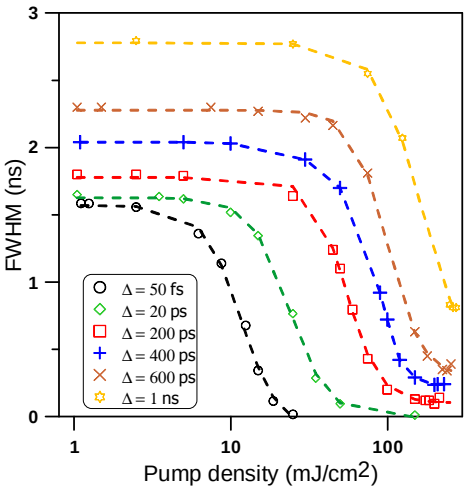

Figure 1. Theoretical FWHM of the pulses as a function of the pump density. (○) Δ = 50 fs, (◊) Δ = 20 ps, (□) Δ = 200 ps, (+) Δ = 400 ps, (×) Δ = 600 ps, (¤) Δ = 1 ns. The dashed lines are the sigmoidal fits to the data.

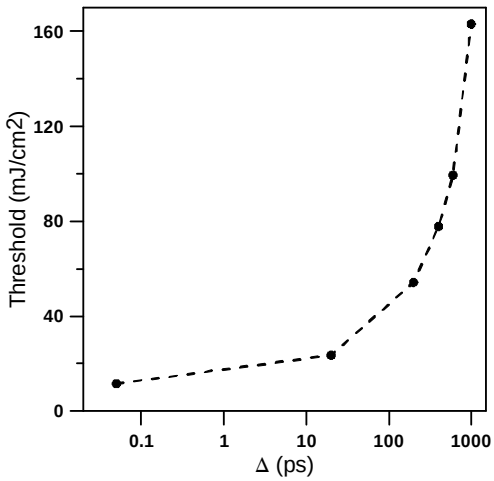

Figure 2. Lasing threshold as a function of the Δ incident pulse. The Δ values are the same as in Figure 1. The dashed line is a guide for the eye.

5. Conclusion

In this work, it is shown that the Crank-Nicholson finite difference method solves satisfactorily the laser rate equations of powder random lasers when a diffusive propagation of light is considered.The method has been applied for one and two-photon excitation. The numerically calculated results in a Rhodamine 6G doped ground powder agree with the corresponding experimental results [5, 7].

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