Time Dependent Theory for Random Lasers

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A model to simulate the phenomenon of random lasing is presented. It couples Maxwell's equations with the rate equations of electronic population in a disordered system. Finite difference time domain methods are used to obtain the field pattern and the spectra of localized lasing modes inside the system. A critical pumping rate $P_c^*$ exists for the appearance of the lasing peaks. The number of lasing modes increases with the pumping rate and the length of the system. There is a lasing mode repulsion. This property leads to a saturation of the number of modes for a given size system and a relation between the localization length $\xi$ and average mode length $L_m$.

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The interplay of localization and amplification is an old and interesting topic in physics research [1]. With promising properties, mirrorless random laser systems are widely studied [2–7] both experimentally and theoretically. Recently, new observations [2] of laserlike emission were reported and showed new interesting properties of amplifying media with strong randomness. First, sharp lasing peaks appear when the gain or the length of the system is over a well-defined threshold value. Although a drastic spectral narrowing has been previously observed [3], discrete lasing modes were missing. Second, more peaks appear when the gain or the system size further increases over the threshold. Third, the spectra of the lasing system is direction dependent, not isotropic. To fully explain such an unusual behavior of stimulated emission in random systems with gain, we are in need of new theoretical ideas.

Based on the time-dependent diffusion equation, earlier work of Letokhov [1] predicted the possibility of lasing in a random system and Zyuzin [5] discussed the fluctuation properties near the lasing threshold. Recently, John and Pang [6] studied the random lasing system by combining the electron number equations of energy levels with the diffusion equation. By using the diffusion approach, it is not possible to explain the lasing peaks observed in the recent experiments [2] in both semiconductor powders and in organic dyes-doped gel films. Another approach which is based on the time-independent wave equations for the random gain media can go beyond the diffusive description [7–11], but is useful only in determining the lasing threshold [12]. To fully understand the random lasing system, we have to deal with time-dependent wave equations in random systems by coupling Maxwell’s equations with the rate equations of electron population within a semiclassical [13,14] theory.

In this paper, we introduce a model by combining these semiclassical laser theories with Maxwell’s equations. By incorporating a well-established FDTD (finite-difference time-domain) [15] method we calculate the wave propagation in random media with gain. Because this model couples electronic number equations at different levels with field equations, the amplification is nonlinear and saturated, so stable state solutions can be obtained after a long relaxation time. The advantages of this FDTD model are obvious, since one can follow the evolution of the electric field and electron numbers inside the system. From the field distribution inside the system, one can clearly distinguish the localized modes from the extended ones. One can also examine the time dependence of the electric field inside and just outside the system. Then after Fourier transformation, the emission spectra and the modes inside the system can be obtained.

Our system is essentially a one-dimensional simplification of the real experiments [2,3]. It consists of many dielectric layers of real dielectric constant of fixed thickness, sandwiched between two surfaces, with the spacing between the dielectric layers filled with gain media (such as the solution of dye molecules). The distance between the neighboring dielectric layers is assumed to be a random variable. The overall length of the system is $L$.

Our results can be summarized as follows: (i) As expected for periodic and short ($L < \xi$, $\xi$ is the localization length) random system, an extended mode dominates the field and the spectra. (ii) For either strong disorder or the long ($L \gg \xi$) system, we obtain a low threshold value for lasing. By increasing the length or the gain more peaks appear in the spectra. By examining the field distribution inside the system, one can clearly see that these lasing peaks are coming from localized modes. (iii) When the gain or the pumping intensity increases even further, the number of lasing modes do not increase further, but saturate to a constant value, which is proportional to the length of system for a given randomness. And (iv) the emission spectra are not the same for different output directions which show that the emission is not isotropic. These findings are in agreement with recent experiments [2] and also make new predictions. We want to point out that our model is 1D unlike the experiments which are done in 3D samples. However, the experimental results are strongly dependent on the shape of the excitation area [2]. Sharp lasing peaks are observed when the excitation area is...
The equations giving the number of electrons on every level can be expressed as follows:

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\begin{align*}
\frac{dN_3(x,t)}{dt} &= P_r N_0(x,t) - \frac{N_3(x,t)}{\tau_{32}} \\
\frac{dN_2(x,t)}{dt} &= \frac{N_3(x,t)}{\tau_{32}} + \frac{E(x,t)}{\hbar \omega_{\alpha}} \frac{dP(x,t)}{dt} - \frac{N_2(x,t)}{\tau_{21}} \\
\frac{dN_1(x,t)}{dt} &= \frac{N_2(x,t)}{\tau_{21}} - \frac{E(x,t)}{\hbar \omega_{\alpha}} \frac{dP(x,t)}{dt} - \frac{N_1(x,t)}{\tau_{10}} \\
\frac{dN_0(x,t)}{dt} &= \frac{N_1(x,t)}{\tau_{10}} - P_r N_0(x,t),
\end{align*}
\]

where \(\frac{E(x,t)}{\hbar \omega_{\alpha}} \frac{dP(x,t)}{dt}\) is the induced radiation rate or excitation rate depending on its sign.

To excite the system, we must introduce sources into the system [17]. To simulate the real laser system, we introduce sources homogeneously distributed in the system to simulate the spontaneous emission. We make sure that the distance between the two sources \(L_s\) is smaller than the localization length \(\xi\). Each source generates waves of a Lorentzian frequency distribution centered around \(\omega_{\alpha}\), with its amplitude depending on \(N_2\). In real lasers, the spontaneous emission is the most fundamental noise [13,14], but generally submerged in other technical noises which are much larger. In our system, the simulated spontaneous emission is the only noise present, and is treated self-consistently. This is the reason for the small background in the emission spectra shown below.

There are two leads, both with a width of 300 nm, at the right and the left sides of the system, and at the end of the leads we use the Liao method [15] to impose absorbing-boundary conditions (ABC). In the FDTD calculation, we discretize both the space and time. The discrete time step and space steps are chosen to be \(10^{-17}\) s and \(10^{-9}\) m, respectively. In the FDTD scheme the boundary conditions for the field at the interfaces between the two media are automatically satisfied, since we are numerically solving Maxwell’s equations. So for a given random configuration, based on the previous time steps we can calculate the next time step \((n + 1)\text{ step}\) values. First we obtain the \(n + 1\) time step of the electric polarization density \(P\) by using Eq. (1), then the \(n + 1\) step of the electric and magnetic fields are obtained by Maxwell’s equations, and at last the \(n + 1\) step of the electron numbers at each level are calculated by Eq. (2). The initial state is that all electrons are on the ground state, so there is no field, no polarization, and no spontaneous emission. Then the electrons are pumped and the system begins to evolve according to the above equations.

We have performed numerical simulations for periodic and random systems. First, for all the systems, a well-defined lasing threshold exists. As expected, when the randomness becomes stronger, the threshold intensity decreases because localization effects make the paths of waves propagating inside the gain medium much longer.
For a periodic or short \((L < \xi)\) random system, generally only one mode dominates the whole system even if the gain increases far above the threshold. This is due to the fact that the first mode can extend in the whole system, and its strong electric field can force almost all the electrons of the upper level \(N_2\) to jump down to the \(N_1\) level quickly by stimulated emission. This leaves very few upper electrons for stimulated emission of the other modes. In other words, all the other modes are suppressed by the first lasing mode even though their threshold values are only a little bit smaller than the first one. This phenomenon also exists in common homogeneously broadening lasers [13].

For long \((L \gg \xi)\) random systems, richer behavior is observed. First we find that all the lasing modes are localized and stable around their localization centers after a long time. Each mode has its own specific frequency and corresponds to a peak in the spectrum inside the system. When the gain increases beyond the threshold, the electric field pattern (see Fig. 1a) shows that more localized lasers correspond to a peak in the spectrum inside the system. In Fig. 1c, the time dependence of the averaged \(A_E\) over all the spatial points is shown. \(I_e\) is obtained by taking the Fourier transform of the spatial averaged \(A_E(t)\).

The exact position of the lasing peaks depends on the random configuration. Notice that the lasing peaks are much narrower than the experimental ones [2]. This is due to the 1D nature of our model. In the present case only two escaping channels exist, so it’s more difficult for the wave to get out from the system which has a higher quality factor. When the gain is really big, we find the number of lasing modes will not increase any more, so a saturated number \(N_m\) of lasing modes exists for the long random system. In Fig. 2, we plot the spectral intensity vs the wavelength for different input transitions (or equivalently pumping rates). Notice these results are in qualitative agreement with the experimental results shown in Fig. 2 of the paper of Cao et al. [2].

These multilasing peaks and the saturated-mode-number phenomena are due to the interplay between localization and amplification. Localization makes the lasing mode strong around its localization center and exponentially small away from its center so that it only suppresses the modes in this area by reducing \(N_2\). When a mode lases, only those modes which are far enough from this mode can lase afterwards. So more than one mode can appear for a long system and each mode seems to repel each other. Because every lasing mode dominates a certain area and is separated from other modes, only a limited number of lasing modes can exist for a finite long system even in the case of large amplification. We therefore expect that the number of surviving lasing modes \(N_m\) should

FIG. 1. (a) The amplitude of the electric field \(A_E\) in units of \(V/m\) vs the position \(L\) in the system, (b) the density of electrons in the \(N_2(N_1)\) in units of \(1/m^3\) levels vs \(L\), (c) \(A_E\) in units of \(V/m\) average over length vs \((t - t_i)/t_0\), where \(t_0 = 2.78 \times 10^{-17}\) s and \(t_i = 7 \times 10^{-10}\) s, and (d) the spectra \(I_e\) in units of \(V/m\) vs the wavelength for a 80 cell system with \(W = 1.4\), \(N_0 = 5.5 \times 6.02 \times 10^{23}\) m\(^{-3}\), and \(P_r = 1 \times 10^7\) s\(^{-1}\).

FIG. 2. The spectral intensity in arbitrary units vs the wavelength for the 80 cell system with \(W = 1.4\) for different pumping rates \(P_r\). \(P_r\) in units of s\(^{-1}\) is (a) \(10^0\), (b) \(10^2\), and (c) \(10^{10}\). To be able to plot all the curves in one figure, we have multiplied (a), (b), and (c) by \(10^{-3}\), \(10^{-5}\), and \(10^{-7}\), respectively, and shifted them apart.
be proportional to the length of the system $L$ when the amplification is very large. Since the “mode-repulsion” property is coming from the localization of the modes, we expect that the average mode length $L_m = L/N_m$ should be proportional to localization length $\xi$ too. In Fig. 3, we plot $N_m$ vs the length of the systems $L$ when we increase the length from 80 cells to 320 cells and keep all other parameters the same. In Fig. 3, we also plot the average mode length $L_m$ vs the localization length $\xi$ when we change the random strength $W$ for a 320 cell system. The localization lengths are calculated using the transfer-matrix method by averaging 10000 random configurations. These results confirm that indeed $N_m \propto L$ and $L_m \propto \xi$. It will be very interesting if these predictions can be checked experimentally.

The emission spectra at the right and left side of the system are quite different. This can be explained from the field pattern shown in Fig. 1a. Notice the localized modes are not similar at both sides of the system. This is the reason for this difference in the output spectrum.

The nonisotropic output spectra of real 3D experiments [2] might be explained by assuming that every localized mode has its intrinsic direction, strength, and position, and the detected output spectra in experiments at different directions are the overlap of contributions from many modes. So generally they should be different.

In summary, by using a FDTD method we constructed a random lasing model to study the interplay of localization and amplification. Unlike the time-independent models, the present formulation calculates the field evolution beyond the threshold amplification. This model allows us to obtain the field pattern and spectra of localized lasing modes inside the system. For random systems, we can explain the multipeaks and the nonisotropic properties in the emission spectra, seen experimentally. Our numerical results predict the mode-repulsion property, the lasing-mode saturated number, and average mode length. We also observed the exchange of energy between the localized modes which is much different from common lasers and this is essential for further research of mode competition and evolution in random laser.

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