

**Coupling, competition, and stability of modes in random lasers**

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We studied analytically and numerically the complex properties of random lasing modes. Mode repulsion in frequency domain for inhomogeneously broadened gain media was confirmed by our numerical results. We constructed a coupled-mode model to explain the synchronized lasing behavior for modes whose frequency difference is less than the homogeneous gain width. The stability of coupled modes was investigated. The effective competition coefficient  $C_e$  for two modes with both gain competition and field coupling is obtained analytically. In our numerical experiments, we also found the coupled oscillations of two lasing modes. From the analytical derivation, we demonstrated that such oscillations could reveal the field-coupling strength between the random modes.

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**I. INTRODUCTION**

Over the past decade, random lasers received much attention of both theoretical and experimental groups.<sup>1-18</sup> In addition to many potential applications, the study of random lasers is important from the fundamental physics point of view. It bridges two basic physics branches, laser physics (electromagnetic waves in gain systems) and Anderson localization (wave propagation in random systems). Random laser not only becomes a new member of laser family,<sup>1,2</sup> but also provides a new path to study localization phenomena.<sup>6,8</sup> Unlike the conventional lasers, the modes of random lasers are formed by random scattering instead of designed reflection. Because random lasing modes come from the eigenstates of disordered systems,<sup>6</sup> they open a special door to study the interplay between localization and amplification.<sup>15</sup>

Although random lasing modes have been studied<sup>2,5,6,8</sup> and the interaction between them been observed,<sup>5,8</sup> a detailed theoretical study is still missing. The interaction between the random lasing modes provides us a rare chance to quantitatively study the properties of random modes. The information obtained from the studies can reveal the fundamental specialty of random lasers. We know it is hard to extract such detailed information from the transport studies of passive random systems for following reasons; (i) The physical quantities in the transport measurement, such as transmission, reflection, and density of states, are *averaged* over many modes of the random systems; (ii) it is very difficult to excite the modes deep *inside* a random system with an optical field incident from outside. Other reasons, such as the short lifetime of excited random modes, also make the experimental and theoretical study difficult. These difficulties, however, can be avoided in random lasers. First of all, only a few modes lase, thus the interaction between them is not averaged anymore. Second, the modes inside the random system can be excited, for example, by electric pumping. Finally, the pumping parameters, such as the strength, the

time length, and the spatial area, can be controlled, so that excited mode can survive enough time length for observing the interaction between the lasing modes. Real and numerical experiments show that the interactions between random lasing modes could be observed, e.g., the mode repulsion in real space and spectral domain, the complex dynamic processes of coupled modes.<sup>5,8</sup>

The interaction between random modes in active random media can be separated into two kinds.

(i) Direct coupling of electromagnetic (EM) field between modes, such as the field leaked from one lasing mode can be absorbed by other modes. Because of the finite size of random lasing systems, the eigenfrequency of a mode (strictly should be called quasimode) is a complex number, whose imaginary part describes the decay rate. Because the eigenfrequencies are complex values, the eigenmodes are not orthogonal to each other. This leads to a linear field coupling between the modes (quasimodes). Such mode coupling could be strong if two modes are spatially and spectrally close to each other.

(ii) Competition of gain between modes which are spatially overlapped, also called cross saturation in laser physics. The process can be explained as that the local depletion of excited electrons by one lasing mode will suppress other mode to lase.

The physics importance of these two kinds of interaction between modes are obvious. Because the coupling between the random modes can tell us how the field leaks from one mode to another one, it is related to the propagation channels of EM field in random system. Hence, a clear physical picture of such coupling is essential for understanding wave localization. The competition of gain is related to the local interaction of the EM field with excited electrons (or other active particles). Such competition can be regarded as an indirect interaction between random modes through the active media. For two modes that are close in frequency and in space, the coupling and competition could be strong. Such

interactions may strongly change the properties of lasing modes, e.g., they influence which mode would lase, the dynamic behavior of the lasing modes, the spectral property of random laser, etc. A theoretical study of mode interactions would allow us to understand thoroughly the physics behind random lasing phenomena.

In our theoretical discussion, reabsorption and reemission of photons are ignored with the approximation that the electronic lifetime at lower-lasing level is very short. Because the photons from one lasing mode can be reabsorbed by electrons at lower-lasing level and reexcited electrons can emit photons into other modes, so the process will cause an extra coupling between modes. But in our four-level electronic model, if the lifetime of lower-lasing level is very short (it is true generally), such effects could be very weak.

Recently, some experimental results<sup>8</sup> have shown the complex behaviors of random lasing modes when there are coupling and competition between them in inhomogeneous active media. We know, that the gain competition of modes in homogeneously broadened gain material causes the modes spatially to repel each other and their distance is related to the localization length if the frequency is in the localized regime. This was first predicted theoretically,<sup>5</sup> then observed experimentally.<sup>17</sup> But the inhomogeneous broadening of gain material adds one more dimension to mode competition. Because in inhomogeneous gain material, the active atoms are separated into several subgroups. The gain of each subgroup is homogeneous and is centered around a certain frequency. The central frequencies of all subgroups are evenly distributed in frequency domain, but the weights of different subgroups are different. Such subgroups (the source of inhomogeneous broadening) in real material could be caused by many physical effects, such as the Doppler effect of the excited gas atoms or the nearly degenerated energy levels in dye molecules. So the mode competition of gain exists not only in spatial domain, but also in spectral domain (different subgroups). In the experiments,<sup>8</sup> the gain material (rhodamine 640 dye in glassy host) was inhomogeneously broadened, and only a *small* area (smaller than the typical mode size) was pumped. Thus all lasing modes should be spatially overlapped. In other words, the strong spatial competition is locked in the experimental condition, so that only one mode would lase and suppress all others if the gain was homogeneous. The experimentally observed coexistence of lasing modes can only be explained by the fact that the inhomogeneous broadening of gain gives them a new dimension (subgroups) to survive (lase simultaneously). At a high pumping intensity (when a single subgroup can support a lasing mode), lasing modes are found to be regularly spaced in frequency. That is an indication of mode repulsion in the spectral domain. Every lasing mode depletes only a subgroup of excited molecules. Each subgroup of excited molecules could be regarded as a homogeneously broadened gain source at certain frequency, and it can only support one lasing mode around that frequency. The frequency spacing of lasing modes corresponds to the homogeneous linewidth of the subgroup molecules. We will see that such phenomena really also appear in our numerical simulation in a similar condition.

Despite the fact that most lasing modes are spectrally separated in the random laser with inhomogeneously broadened gain spectrum, two special cases are found experimentally. One is the observation of two lasing modes whose frequency spacing is less than the homogeneous linewidth of the gain material. Their field strengths are similar. The temporal measurement reveals that the evolution of these two modes is synchronized, i.e., lasing starts and stops at the same time. This obviously tells us that there exists another mechanism which can balance the mode competition. This mechanism is the field coupling between the two modes, namely, the EM field leaked from one mode is absorbed by another. When the frequencies of two modes are close, their coupling could be very strong. The other case is that two modes close in frequency are coupled, but their field strengths are quite different. Their lasing period is different. The stronger mode (called main mode) lases longer than the weaker one (called side mode). Both phenomena are also observed in the random laser with homogeneously broadened gain spectrum.

In our numerical simulations, we found additional phenomena and dynamic processes of coupled modes that are not observed experimentally. For example, we observed the coupled oscillations of two strongly coupled random modes. To explain all these results, we need a theoretical model. In this paper, we presented quantitative treatment of these phenomena by semiclassical laser theories.<sup>19,20</sup> Due to the complexity of the active random systems, we have to make some approximation, which will be discussed in the later sections. Our goal is to find the physical mechanisms behind all the phenomena described earlier. This paper is organized as follows. Section II contains the numerical results of mode repulsion in frequency domain of inhomogeneously broadened random laser. Based on the solution to the coupled field equations of two modes, we give the theoretical explanation of synchronized lasing behavior of two coupled modes. In Sec. III, stability of two gain-competed and field-coupled modes is studied with rate equations. The criterion of stability is presented. Section IV focuses on the coupled relaxation oscillations of two strongly coupled lasing modes. Theoretically, we find that the frequencies of such oscillations reveal the coupling strength between the random modes. In Sec. V, we summarize the results and emphasize the importance of studying the random lasing modes.

## II. MODE REPULSION AND SYNCHRONIZED LASING

First we checked the experimental observation of mode repulsion in frequency domain of inhomogeneously broadened gain material by our numerical simulation. The inhomogeneously broadened gain spectrum was constructed with numerous homogeneous lines. The inhomogeneous linewidth is 150 THz, 30 times of the homogeneous linewidth  $\Delta\omega = 5$  THz.<sup>5,12</sup> The Maxwell's equations are coupled to the rate equations of electronic populations. Temporal evolution of the electromagnetic field is calculated with the finite difference time domain method. The lasing spectrum is obtained by Fourier transform. For simplicity, we calculated one-dimensional (1D) random lasers which could reveal

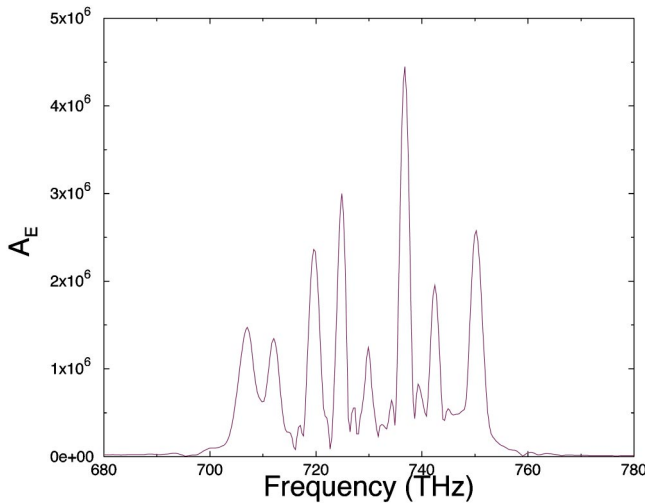


FIG. 1. Numerically calculated lasing spectrum at the pumping rate of  $P_r = 1 \times 10^7 \text{ s}^{-1}$  of a 1D random laser system with inhomogeneous gain material. Almost regularly spaced peaks are shown.

qualitative properties of 2D and 3D random lasers. The behaviors of 2D and 3D random lasers are qualitatively the same. Our 1D random system is made of binary layers with dielectric constants  $\epsilon_1 = 2.5$  and  $\epsilon_2 = 5.29$ . The layer thickness  $a = a_0(1 + W_a\gamma)$ ,  $b = b_0(1 + W_b\gamma)$ , where  $a_0 = 48 \text{ nm}$ ,  $W_a = 0$ ,  $b_0 = 80 \text{ nm}$ ,  $W_b = 0.2$ ,  $\gamma$  is a random value in the range  $[-1, 1]$ . The system consists of 700 pairs of binary layers, the system length  $\sim 100 \mu\text{m}$ . Only the central part of the system with length  $30 \mu\text{m}$  is pumped. Due to the weak randomness, the modes are spatially extended. Thus the lasing modes must overlap with each other and compete for the gain. At high pumping rate, the lasing modes are almost regularly spaced in frequency as shown in Fig. 1. We also repeated the calculation of similar systems with different parameters, and found that the regular spacing of lasing peaks is a common feature of the lasing spectra. The frequency separation of neighboring lasing modes is about 7 THz, which is a little smaller than the theoretical value  $2\Delta\omega = 10 \text{ THz}$ .<sup>21</sup> We note that the frequency spacing between adjacent lasing modes is much larger than the average frequency spacing between eigenmodes in the random systems. This indicates that lasing in most eigenstates is suppressed because their frequencies are too close to one of the lasing modes which has depleted the gain in one subgroup of excited molecules. Cross saturation of gain allows only one mode to lase within a homogeneous line of the gain material.

However, we do observe two kinds of exceptions as in experiments. Next we will focus on the first kind. We observe that two lasing modes are spatially overlapped and the frequency separation between them is less than the homogeneous linewidth. Thus they should interact with the same subgroup of excited molecules, and the gain competition would suppress lasing in one of them if there were no other interaction. The facts that they lase simultaneously and their field strengths are similar suggest that there must be additional interactions between them. This additional interaction is the field coupling between the modes. It also exists in

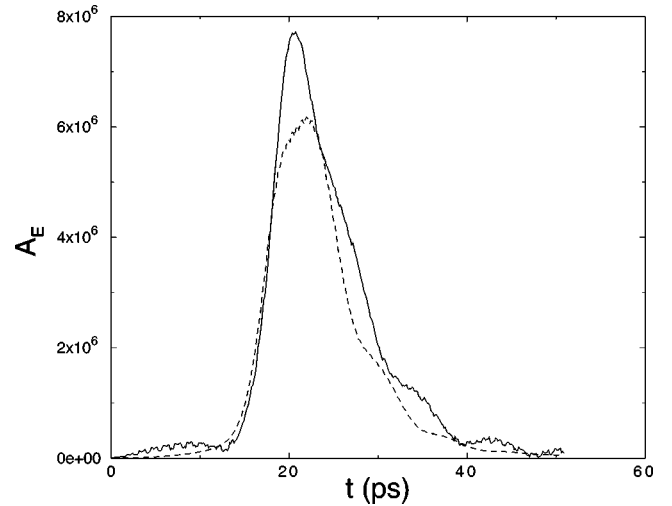


FIG. 2. The field amplitudes  $A_E$  of two lasing modes vs time  $t$ . The synchronized lasing of two modes are shown.

modes of random lasers with homogeneously broadened gain spectrum.

Numerically, we simulated such phenomena in a homogeneous gain system. The 1D random system consists of 100 pairs of binary layers with the randomness  $W_a = 0.1$  and  $W_b = 0.6$ . The small system size allowed us to calculate many random configurations and find one with two coupled modes. The pumping process starts at  $t = 0$  and stops at  $t = 15 \text{ ps}$ . At a pumping rate  $P_r = 4 \times 10^6 \text{ s}^{-1}$ , the spectrum has two lasing peaks with frequency separation 0.8 THz. Figure 2 shows the temporal evolution of the amplitude of the electric field  $A_E$  for these two lasing modes. Laser pulses in these two modes rise at the same time. The pulse duration is nearly identical. These results agree with the experimental data.

The next question is whether such synchronized lasing modes can be obtained in theoretical analysis. Our answer is yes. Because the system, including the EM fields and electron populations for two modes, is complex, we would end up with a large number of coupled differential equations if we do not make any approximation or simplification. Strictly speaking, when two modes are close in frequency, the EM field equations should be used to explain the detailed coupling and competition between them. For the field equations, we separate the space and time variables, and assume the equations for the spatial part are solved. The eigenmodes and eigenfrequencies are already obtained. We only need to solve the time-dependent part. With the field equations for modes, we can get the instantaneous interaction between the modes. However, the field equations (even scalar field equations) could be too hard to solve sometimes, especially when they are coupled with the rate equations for electrons. In this paper, we will use the scalar field equations to study the synchronization of two coupled lasing modes, but use the photon rate equations to investigate their stabilities. Such simplification is reasonable because the synchronization is related to the phases of mode oscillations which could only be studied with the field equations, while the stability is the time-averaged behavior of the lasing modes. These methods



are commonly used in the laser physics<sup>21</sup> when the dynamic processes are studied.

Let us consider two modes  $A$  and  $B$ . Their frequencies  $\omega_A$  and  $\omega_B$  are close, and they are spatially overlapped. Their quality factors are slightly different. The coupled field equations are

$$\begin{aligned} \frac{d^2 E_i(t)}{dt^2} + \gamma_i \frac{dE_i(t)}{dt} + \omega_i^2 E_i(t) \\ = -\frac{1}{\varepsilon} \frac{d^2 P_i(t)}{dt^2} + \gamma_{ij} \left( \frac{V_j}{V_i} \right)^{1/2} \frac{dE_j(t)}{dt}. \end{aligned} \quad (1)$$

The subscript  $i, j$  can be 1 (for mode  $A$ ) or 2 (for mode  $B$ ).  $E_i(t)$  is the electric field. The polarization  $P(t)_i$  can be separated into two parts:  $P_i(t) = \varepsilon_0(\chi' + i\chi'')E_i(t)$ , the imaginary part is the source of optical gain.  $\omega_i$  is the eigenfrequency of the passive system.  $V_i$  is the spatial volume of the mode;  $\varepsilon$  is the average dielectric constant.  $\gamma_i = \omega_i/Q_i$  is the decay rate of the cavity, where  $Q_i$  is the quality factor.  $\gamma_{ij}$  is the field-coupling constant, and it can be expressed in the form  $\gamma_{ij} = G_{ij}/C$ , where  $G_{ij}$  is the external-source conductivity and  $C$  is the capacity in the effective circuit model.

Equation (1) is widely used in the dynamic study of a laser cavity with external source. Generally, the coupling constant  $\gamma_{ij}$  is proportional to  $\int E_i(x)E_j(x)dx$  which includes both spatial and spectral overlap information of two modes. In the random laser, if the modes are localized,  $E_1 \propto \exp|x-x_1|/\xi_1$  and  $E_2 \propto \exp(-|x-x_2|)/\xi_2$ . Then  $\gamma_{ij}$  represents the overlap of the tails of the two localized states. Its value is proportional to  $\exp(-|x_2-x_1|/\xi)$ , where  $\xi$  is the larger one in  $\xi_1$  and  $\xi_2$ . Hence, the coupling can be neglected when the spatial distance between the two modes is much larger than  $\xi_1$ . In other words, the coupling constant is a parameter that reveals the spatial overlap of the localized states. If the central frequencies of two modes are far away, the integral  $\int E_i(x)E_j(x)dx$  is almost zero; but if they are near to each other so that the spectral distance is comparable with the widths of peaks (the tails of peaks are overlapped) the coupling constant could be large enough to have physical effects. The last term in Eq. (1) can be thought of as an external force to the mode  $i$ . The coupling effect depends on the frequency difference between the two modes too. Only when the frequency  $\omega_j$  of the external force is close to the mode eigenfrequency  $\omega_i$  (in resonance), the effect of external force is dramatically enlarged. Both conditions, being spatially and spectrally close to each other, are needed for the strong-coupling effect. On the other hand, it is known in Anderson localization theory that such frequency-close modes *should* be spatially separated far away so that they are orthogonal to each other. How can *both* conditions be satisfied? As we discussed above, the orthogonality of localized modes is true for infinite-long random passive systems (cold cavities), but in our real *finite* random *active* systems, there are two reasons for modes to overcome the orthogonality and satisfy both conditions. First, the modes in a finite system are actually quasimodes; two quasimodes are not strictly orthogonal to each other because their resonant spectral peaks are over-

lapped at tails. We can think that such two localized lasing modes are two cavities connected by a small window. If the window is not large, two lasing modes and their lasing frequencies are still well defined. Actually, the distance between two coupled modes is a little larger than the localization lengths of the modes, but the coupling is strong enough to generate physical effects. Second, the saturable gain adds a small nonlinear part [a complex value, see  $P$  in Eq. (1)] to the dielectric coefficient. Such an active nonlinearity can make two orthogonal passive modes to couple to each other, and can lead to more complicated behaviors (i.e., the phase lock) if its value is large enough. In our numerical configurations, the strong-coupling phenomena are pretty rare to be observed, just as those in experiments.<sup>2</sup> Next we will see that the coupling effects are observable, and such effects provide a different way to study the interaction between localized modes in real finite systems.

The general solutions of coupled Eq. (1) are very hard to obtain, but we can get approximate solutions under certain conditions. To simplify our derivation we need to make some approximations. (i) We neglect the gain saturation effect; (ii) we assume the quality factors of modes  $A$  and  $B$  are not very different; (iii) we assume mode  $A$  reaches the lasing threshold first and its field is much stronger than that of mode  $B$ , so that the field output from mode  $A$  influences mode  $B$ , but the reverse influence of mode  $B$  on mode  $A$  is weak and ignored. After the pumping starts, mode  $A$  reaches the lasing threshold first. The field of mode  $A$  increases quickly,  $E_1 \propto f(t)$  [ $f(t)$  is an exponentially growing function if mode  $A$  does not interact with mode  $B$ ]. Meanwhile, mode  $B$  is close to (but still below) the lasing threshold. The field equation for mode  $B$  is similar to that of a driven oscillator:

$$\begin{aligned} \frac{d^2 E_2(t)}{dt^2} + \gamma'_2 \frac{dE_2(t)}{dt} + \omega'_2 E_2(t) \\ = -\gamma_{12} \left( \frac{V_a}{V_2} \right)^{1/2} (i\omega_a) E_1^0 f(t) \exp(-i\omega_1 t). \end{aligned} \quad (2)$$

$\gamma'_2 = \gamma_2 - (\varepsilon_0/\varepsilon)\omega_2^2\chi''$  is the total loss which is very small because mode  $B$  is near threshold.  $\omega'_2$  is the frequency of mode  $B$  when we include the pulling effect of the gain medium. Because frequencies of the two modes are close, there is resonant absorption of the field from mode  $A$  by mode  $B$ . Suppose that  $f(t)$  changes much slower in the lasing process of mode  $B$ , because the energy of mode  $A$  is absorbed by mode  $B$  resonantly. The solution to Eq. (2) has the form<sup>22</sup>

$$E_2(t) \sim \gamma_{12} \left( \frac{V_1}{V_2} \right)^{1/2} t f(t). \quad (3)$$

The factor proportional to time  $t$  is from resonant absorption. We can see that the field of mode  $B$  can increase *faster* than that of mode  $A$  although mode  $B$  lases after mode  $A$ . We need to point out that at resonance, the instantaneous energy flow direction is determined by the phase of the EM field (i.e., whether it does positive or negative work in a period), not by the field strength. Because the phase change could be slower than the field strength change, there could be a net energy

flow from mode  $A$  to mode  $B$  even when the field strength of mode  $B$  is the same as mode  $A$ . This can explain our numerical result that the field of the later lasing mode can be stronger than that of the first lasing mode in the cobuildup process. The process also can be understood as that mode  $A$  provides additional pumping to mode  $B$  (while mode  $B$  results in additional loss of mode  $A$ ). The net energy flow from mode  $A$  to mode  $B$  through photon hopping accelerates the lasing process in mode  $B$ . If the quality factors of the two modes are not very different, mode  $B$  will start to lase and its field strength may even catch up with that of mode  $A$  while mode  $A$  still builds up. That is why we observe the two modes lase almost at the same time. Similarly, when the pumping stops, the mode that decays faster will resonantly absorb energy from the slower decaying one. Then the former's decay is slowed down while the latter's decay becomes faster. As a result, the two modes stop lasing at the same time too, as shown in Fig. 2.

Finally, we give some explanation of the second kind of coupled modes. In the second kind, one mode has much higher quality factor than the other. They are called the main mode and the side mode. The side mode is observable in the spectrum because of an enhanced effect. When the difference between the main mode frequency  $\omega_m$  and the side mode frequency  $\omega_s$  is equal to the relaxation-oscillation frequency  $\omega_{sp}$  of the main mode, the EM field of the side is greatly enhanced. This is another kind of resonance effect, and it can be derived from the photon rate equations including the gain saturation. Because this effect has been well studied in laser physics,<sup>19</sup> we will not discuss the details here.

### III. STABILITY OF TWO COUPLED-COMPETING RANDOM MODES

Our numerical study indicates that the lasing of two modes, which are spatially overlapped and close in frequency, can be stable. In principle, when two modes are spatially overlapped the gain competition would allow only one mode to lase. However, in the presence of field coupling between the two modes, second mode could start lasing. How both modes can be stable in the lasing process is still a question. To confirm our numerical results, we need theoretical derivation to understand the stability of these random lasing modes. To simplify our derivation, we solved the photon rate equations. We denoted the two modes  $A$  and  $B$  with the subnotes 1 and 2:

$$\frac{dn_1}{dt} = \frac{\gamma_{g1}n_1}{1 + \kappa_{11}n_1 + \kappa_{12}n_2} - \gamma'_1n_1 + \gamma'_{12}(n_2 - n_1) \quad (4)$$

$$\approx \gamma_{g1}n_1(1 - \kappa_{11}n_1 - \kappa_{12}n_2) - \gamma'_1n_1 + \gamma'_{12}(n_2 - n_1), \quad (5)$$

$$\frac{dn_2}{dt} = \frac{\gamma_{g2}n_2}{1 + \kappa_{22}n_2 + \kappa_{21}n_1} - \gamma'_2n_2 + \gamma'_{21}(n_1 - n_2) \quad (6)$$

$$\approx \gamma_{g2}n_2(1 - \kappa_{22}n_2 + \kappa_{21}n_1) - \gamma'_2n_2 - \gamma'_{21}(n_1 - n_2). \quad (7)$$

$\gamma_{g1}$  is amplification rate of mode  $A$ .  $\gamma'_1$  is the field damping rate of mode  $A$  caused by leakage to the environment and other modes except mode  $B$ .  $\gamma'_{12}$  is the spatial-temporal average of the coupling constant between modes  $A$  and  $B$ .  $\kappa_{11}$  is self-saturation parameter of mode  $A$ , and  $\kappa_{12}$  is cross-saturation parameter which includes the information of gain competition. We have similar definition of  $\gamma_{g2}$ ,  $\gamma'_2$ ,  $\gamma'_{21}$ ,  $\kappa_{11}$ , and  $\kappa_{21}$  for mode  $B$ . We make two approximations: (i) the self-saturation and cross-saturation effects are weak; (ii) the photon hopping current is determined by the difference of photon density. The first approximation is widely used in the laser systems. For the second one, as we discussed above, the field coupling of two modes by photon hopping could be more complex if the resonance effect is included. However, we can suppose this is the averaged effect over a time interval that is much longer than the period of the field.

The physical meaning of cross saturation could be more complex in a random laser than in a conventional laser. If the randomness is very weak (the system is almost homogeneous one), the gain saturation results in the grating effect. The cross-saturation parameter  $\kappa_{ij}$  could be much larger when the frequencies of two modes are close. In general cases of random lasers whose dielectric randomness is much larger than the grating effect (the grating effect will not change the distribution of mode field much),  $\kappa_{ij}$  is  $\propto \int |E_j(x)|^2 |E_i(x)|^2 dx$ . Very different from  $\gamma_{ij}$ ,  $\kappa_{21}$  is proportional to the overlap of intensity of modes, so it is almost independent of the frequency difference between modes. If the modes are localized states, the value of  $\kappa_{ij}$  will strongly depend on the distance between the centers of the two modes.

Following the original notations in laser physics,<sup>20</sup> we can rewrite Eqs. (5) and (7) as

$$\frac{dn_1}{dt} = \left[ \alpha_1 - \beta_1 n_1 - \left( \theta_{12} - \frac{\gamma'_{21}}{n_1} \right) n_2 \right] n_1, \quad (8)$$

$$\frac{dn_2}{dt} = \left[ \alpha_2 - \beta_2 n_2 - \left( \theta_{21} - \frac{\gamma'_{12}}{n_2} \right) n_1 \right] n_2. \quad (9)$$

$\alpha_i = \gamma_{gi} - \gamma'_i - \gamma_{ij}$ ,  $\beta_i = \gamma_{gi}\kappa_{ii}$ ,  $\theta_{ij} = \gamma_{gi}\kappa_{ij}$ . If we set the right sides of the above equations to be zero, we obtain the "zero-gain" curves for the two modes in the  $n_1$  vs  $n_2$  plane. We can see from Eqs. (8) and (9) that the coupling terms change the zero-gain curves for the two modes from straight lines to hyperbolic lines. To show the effect of mode coupling on the stability of competing modes, we choose the following parameters:  $\alpha_1 = 1.1$ ,  $\beta_1 = 0.4$ ,  $\theta_{12} = 0.8$ ,  $\alpha_2 = 0.9$ ,  $\beta_2 = 0.3$ ,  $\theta_{21} = 0.7$ ,  $\gamma'_{12} = \gamma'_{21} = 0.35$ . The zero-gain curves are plotted in Fig. 3. First we set the field-coupling terms  $\gamma'_{ij} = 0$ , i.e., no coupling of modes. The zero-gain lines for the two modes should be straight lines, which are shown by the dotted lines in Fig. 3. The gain-competition factor which is defined as  $C = \theta_{12}\theta_{21}/\beta_1\beta_2 > 1$  is shown in Fig. 3 by the angle  $AWB$  larger than  $\pi$  from original side. So that the two modes cannot be stable simultaneously. Only one mode, either mode  $A$  (at point  $A$  in Fig. 3) or mode  $B$  (at point  $B$  in Fig. 3), can lase. Next the field-coupling terms are

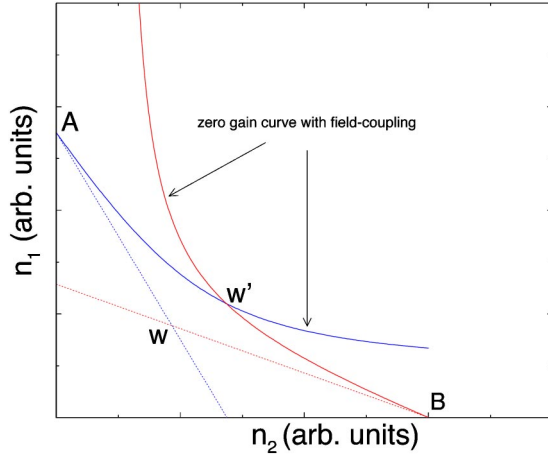


FIG. 3. Zero-gain curves to show the stable property of two modes with  $iZ$ interaction. The solid lines are the zero-gain curves for two modes in the presence of both gain competition and field coupling. The dotted lines are the zero-gain curves without field coupling.

turned on. In the presence of field coupling, the effective competition parameter near the crossing point of two hyperbolic lines is

$$C_e = (\theta_{12} - \gamma'_{12}/n_1)(\theta_{21} - \gamma'_{21}/n_2)/\beta_1\beta_2. \quad (10)$$

The value of  $C_e$  could be smaller than 1 while  $C$  is larger than 1. As shown in Fig. 3, the angle (from original side) around the cross point  $W'$  is smaller than  $\pi$ . Then the two modes can be stable, i.e., both modes lase.

The above derivation shows that the stability for two lasing modes depends on the value of their effective competition factor  $C_e$  at the crossing point, larger or smaller than 1. The next question is the following: how can the two modes reach the crossing point of their zero-gain curves? In other words, how can the two modes reach their stable working condition? As we discussed in the preceding section, the resonance absorbing of the energy between the two modes can help them get to the stable lasing condition. If the effective competition factor  $C_e < 1$  both modes can be stable simultaneously, if  $C_e > 1$  one mode will be suppressed. We note that the effective competition parameter  $C_e$  depends on the mode-coupling parameters, thus the abnormal stability of random lasing modes is a representation of mode-coupling effect.

#### IV. RELAXATION OSCILLATIONS OF STRONGLY COUPLED MODES

In our numerical study, we also observed the coupled oscillations of two modes. The configuration in the central part of the 1D random system is kept the same as that used in the study of synchronized lasing in Sec. III. We add ten pairs of binary layers at each end of the system. The pumping time is increased from 15 ps to 25 ps. Thus the coupled modes are almost the same as in Sec. III, but their quality factors are larger and their lasing time should be longer. Tracking their field amplitudes in time, we can clearly see the coupled os-

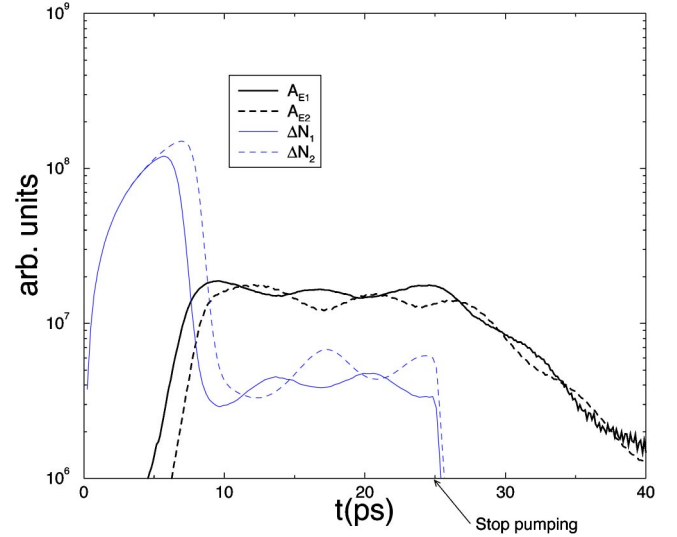


FIG. 4. The coupled oscillations of two lasing modes. Both field magnitudes  $A_E$  and electron population inversions  $\Delta N$  near the two modes' centers are shown.

cillations of the two modes. The phases of their oscillations are  $\pi$  different. The numerical results are shown in Fig. 4. These coupled oscillations are related to the electronic system; namely, the electronic populations also oscillate. For each mode, the phase of electronic oscillation is almost  $\pi$  different from the oscillation of the field.

To explain this complex photon-electron dynamic process, we need to solve the coupled equations for the EM fields and electronic populations of the two modes. The solutions are complicated and provide little physical insight. Next, we will first present a qualitative understanding and then a simplified model. We argue that the oscillations result from strong coupling of two modes. Without coupling, the relaxation of each mode could cause the field and population oscillate independently. When two modes are strongly coupled, their originally independent relaxation oscillations can be coupled and generate new oscillations. With this understanding, we used the model of coupled oscillators:

$$\frac{\partial^2 n_1}{\partial t^2} + \gamma_1 \frac{\partial n_1}{\partial t} + \omega_{o1}^2 n_1 = \xi_{12} n_2, \quad (11)$$

$$\frac{\partial^2 n_2}{\partial t^2} + \gamma_2 \frac{\partial n_2}{\partial t} + \omega_{o2}^2 n_2 = \xi_{21} n_1, \quad (12)$$

where  $\gamma_1$  and  $\gamma_2$  are the damping rates near the stable working condition,  $\omega_{o1}$  and  $\omega_{o2}$  are the relaxation-oscillation frequencies of two modes without coupling.  $\xi_{12}$  and  $\xi_{21}$  are coupling constants which we will explain later.

For the random systems we studied numerically, the damping rates are small, thus the damping terms can be neglected. We obtain the oscillation-frequencies of two strongly coupled modes:

$$\omega_{\pm}^2 = \frac{\omega_1^2 + \omega_2^2}{2} \pm \sqrt{\left(\frac{\omega_1^2 + \omega_2^2}{2}\right)^2 - \omega_1^2 \omega_2^2 + \xi_{12} \xi_{21}}. \quad (13)$$

If  $\omega_{o1} = \omega_{o2} = \omega_{o0}$  and  $\xi_{12} = \xi_{21} = \xi$ , the frequencies of two coupled modes are  $\omega_{\pm} = \sqrt{\omega_{o0}^2 \pm \xi}$ . We see that there should be two eigenoscillations of the coupled modes. One is fast oscillation, the other is slow oscillation. In our case, the slow oscillation frequency is very small compared to the fast one. This means  $\xi \approx \omega_{o0}^2$ . Thus the fast oscillation is what we observed in our numerical experiment. The oscillation frequency  $\omega_{+} \approx \sqrt{2\xi}$ . From the eigenoscillation functions of  $(n_1, n_2)^T$ , we confirm that the fast oscillation corresponds to the two oscillators which have  $\pi$  phase difference. After comparing Eqs. (13) and (1), the physical meaning of  $\xi$  is clear:  $\xi = \omega_{o0} \gamma_{12}$ . Finally we get

$$\omega = \sqrt{2\omega_{o0}\gamma_{12}}. \quad (14)$$

The above result [or the more general one in Eq. (13)] tells us that the coupling strength  $\gamma_{12}$  between random lasing modes can be extracted from the measurement of the coupled-oscillation frequencies. This is a different method to measure the coupling strength of random modes inside the disordered systems.

## V. SUMMARY

In summary, we have studied the interactions between random lasing modes. Despite the fact that some phenomena, such as gain competition, have been observed in conventional laser systems, in random lasers they are still of importance, because they reveal the properties of random modes in the systems. The gain competition of random lasing modes in frequency domain is numerically confirmed for random la-

sers with inhomogeneously broadened gain spectra. For spatially overlapped lasing modes, gain competition results in their repulsion in frequency. Numerical results and analytical derivation reveal the effect of field coupling on random lasing modes. The synchronized lasing of two coupled modes is explained with the field equations. The stability of field-coupled gain-competing random modes is discussed based on the photon rate equations. The criterion of stability is derived, and its relation to the coupling strength is revealed. Moreover, the coupled oscillation of two strongly coupled modes is observed in our numerical simulation. A simplified mode based on coupled-oscillators illustrates that the frequency of the coupled relaxation oscillation depends on the coupling strength. This discovery provides a different method of measuring the coupling strength of random modes. Since the properties of random modes directly affect light transport in disordered systems, the parameters we obtained from the random laser study, e.g., the coupling and cross-saturation parameters of random modes, facilitate further quantitative studies. Although the lasing modes represent only a special subset of eigenmodes in a random system, the understanding of their complex behaviors opens a new window for fully understanding disordered systems.

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- <sup>1</sup>N.M. Lawandy, R.M. Balachandran, S.S. Gomers, and E. Sauvain, *Nature (London)* **368**, 436 (1994); V.S. Letokhov, *Sov. Phys. JETP* **26**, 835 (1968).
- <sup>2</sup>H. Cao, Y.G. Zhao, S.T. Ho, E.W. Seelig, Q.H. Wang, and R.P.H. Chang, *Phys. Rev. Lett.* **82**, 2278 (1999); H. Cao, J.Y. Xu, D.Z. Zhang, S.-H. Chang, S.T. Ho, E.W. Seelig, X. Liu, and R.P.H. Chang, *ibid.* **84**, 5584 (2000).
- <sup>3</sup>S.V. Frolov, Z.V. Vardeny, K. Yoshino, A. Zakhidov, and R.H. Baughman, *Phys. Rev. B* **59**, R5284 (1999).
- <sup>4</sup>D.S. Wiersma, M.P. van Albada, and Ad Lagendijk, *Phys. Rev. Lett.* **75**, 1739 (1995); D.S. Wiersma *et al.*, *Phys. Rev. E* **54**, 4256 (1996).
- <sup>5</sup>X. Jiang and C.M. Soukoulis, *Phys. Rev. Lett.* **85**, 70 (2000).
- <sup>6</sup>X. Jiang and C.M. Soukoulis, *Phys. Rev. E* **65**, 025601(R) (2002).
- <sup>7</sup>M. Patra, H. Schomerus, and C.W.J. Beenakker, *Phys. Rev. A* **61**, 023810 (2000).
- <sup>8</sup>H. Cao, X. Jiang, Y. Ling, J.Y. Xu, and C.M. Soukoulis, *Phys. Rev. B* **67**, 161101 (2003).
- <sup>9</sup>H. Cao, Y. Ling, J.Y. Xu, C.Q. Cao, and P. Kumar, *Phys. Rev. Lett.* **86**, 4524 (2001).
- <sup>10</sup>R.C. Polson, A. Chipouline, and V. Vardeny, *Adv. Mater.* **13**, 760 (2001).
- <sup>11</sup>C. Vanneste and P. Sebbah, *Phys. Rev. Lett.* **87**, 183903 (2001).
- <sup>12</sup>C.M. Soukoulis, X. Jiang, J.Y. Xu, and H. Cao, *Phys. Rev. B* **65**, 041103(R) (2002).
- <sup>13</sup>A.L. Burin, M. Ratner, H. Cao, and S.-H. Chang, *Phys. Rev. Lett.* **88**, 093904 (2002).
- <sup>14</sup>R.C. Polson, M.E. Raikh, and Z.V. Vardeny, cond-mat/0105360 (unpublished).
- <sup>15</sup>V.M. Apalkov, M.E. Raikh, and B. Shapiro, cond-mat/0111047 (unpublished).
- <sup>16</sup>T.Sh. Misirpashaev and C.W.J. Beenakker, *Phys. Rev. A* **57**, 2041 (1998).
- <sup>17</sup>Y. Ling, H. Cao, A.L. Burin, M.A. Ratner, X. Liu, and R.P.H. Chang, *Phys. Rev. A* **64**, 063808 (2001).
- <sup>18</sup>H. Cao, Y. Ling, J. Y. Xu, and A. L. Burin, *Phys. Rev. E* (to be published).
- <sup>19</sup>A. E. Siegman, *Lasers* (University Science Books, Mill Valley, 1986).
- <sup>20</sup>A. Maitland and M. H. Dunn, *Laser Physics* (North-Holland, Amsterdam, 1969), Chap. 9.
- <sup>21</sup>Anthony E. Siegman, *Lasers* (Mill Valley, California, 1986), Chaps. 2, 3, 6, and 13.
- <sup>22</sup>L. D. Landau and E. M. Lifshitz, *Mechanics* (Robert Maxwell, Oxford, 1987).