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Localized random lasing modes and a path for observing localization

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We demonstrate that a knowledge of the density of states and the eigenstates of a random system without gain, in conjunction with the frequency profile of the gain, can accurately predict the mode that will lase first. Its critical pumping rate can also be obtained. It is found that the shape of the wave function of the random system remains unchanged as gain is introduced. These results were obtained by the time-independent transfer matrix method and finite-difference time-domain methods in a one-dimensional model. They can also be analytically understood by generalizing the semiclassical Lamb theory of lasing in random systems. These findings provide a path for observing the localization of light, such as looking for the mobility edge and studying the localized states.

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Localization theory and laser theory were both developed in the 1960s. The propagation of quantum and classical waves in disordered media is well understood [1], while laser physics has been well established [2,3] at the same time. It was always assumed that disorder was detrimental to lasing action. However, Letokhov [4] theoretically predicted the possibility of lasing in a random system, called a ''random laser.'' Only after the experimental observations by Lawandy *et al.* [5], were random laser systems studied intensively [6–19]. Since then, many experiments have been carried out that showed a drastic spectral narrowing [6] and a narrowing of the coherent backscattering peak [7]. Recently, additional experiments [8,9] showed random laser action with sharp lasing peaks. To fully explain such an unusual behavior of stimulated emission in random systems, many theoretical models were constructed. John and Pang [10] studied the random lasing system by combining the electron number equations of energy levels with the diffusion equation. Berger *et al.* [11] obtained the spectral and spatial evolution of emission from random lasers by using a Monte Carlo simulation. Very recently, Jiang and Soukoulis [12,13], by combining a finite-difference time-domain (FDTD) method with interplay of localization and amplification. They obtained [12,13] the field pattern and the spectral peaks of localized lasing modes inside the system. They were able to explain [12,13] the multiplepeaks and the nonisotropic properties in the emission spectra, seen experimentally [8,9]. Finally, the mode repulsion property which gives saturation of the number of lasing modes in a given random laser system was predicted. This prediction was checked experimentally by Cao *et al.* [14].

One very interesting question that has not been answered by previous studies is, what is the form of the wave function in a random laser system? How does the wave function in a random system change as one introduces gain? Does the wave function retain its shape in the presence of gain? Another very interesting point is whether we can predict *a priori* which mode will lase first. What will be its emission wavelength? If we understand these issues, we will be able to design random lasers with the desired emission wavelengths. In addition, we will be able to use the random laser as a tool to study the localization properties of random systems.

In this paper, we explore the evolution of the wave function without and with gain, by the time-independent transfer matrix theory [15–19], as well as by time-dependent theory [12,13]. The emission spectra can also be obtained. In addition, we can also use the semiclassical theory of lasing [2,3] to obtain analytical results for the threshold of lasing, as well as which mode will lase first. This depends on the gain profile, as well as on the quality factor Q of the modes before gain is introduced.

Our system is essentially a one-dimensional simplification of the real experiments [8,9]. It consists of many dielectric layers of real dielectric constant $\varepsilon_2 = 2.56\varepsilon_0$ (ε_0 is the dielectric permeability of free space) of fixed thickness ($b_0 = 100$ nm), sandwiched between two surfaces, with the spacing between the dielectric layers assumed to be a random variable $a_n = a_0(1+W)$, where $a_0 = 200$ nm and W has a random value in the range of $[-0.7, 0.7]$. We choose a 30-cell random system, as the first system of our numerical study. In Fig. 1(a), we present the results for the logarithm of the transmission coefficient as a function of frequency f. These results were obtained by using the transfer matrix techniques introduced in Ref. [15]. Notice that we have three typical resonance peaks (denoted P_1, P_2, and P_3) in the frequency range of 600 to 660 THz. As one can see from Fig. 1(a), the linewidths of the three modes are different. P_3 has the smallest linewidth and therefore the largest Q, while P_1 has the largest linewidth and therefore the smallest Q. We have also numerically calculated the wave functions corresponding to these three peaks and indeed find out that the more localized wave function is the one with the larger Q. All the results above were obtained for the case without gain.

According to the semiclassical theory of laser physics [2,3], we generally use a polarization due to gain $P_{gain} = \varepsilon_0 \chi(\omega) E = \varepsilon_0 [\chi'(\omega) + i\chi''(\omega)] E$ to introduce amplifying medium effects. Both $\chi'(\omega)$ and $\chi''(\omega)$ are proportional to the outside pumping rate P_r and can be expressed by the parameters of the gain material [20].

To determine which peak will lase first, we can again use the time-independent transfer matrix method [see Eq. (4) of Ref. [15]] with a frequency-*independent* gain, which means the width of the gain profile is very large. It is well understood that time-independent theory [15–19] for random la-

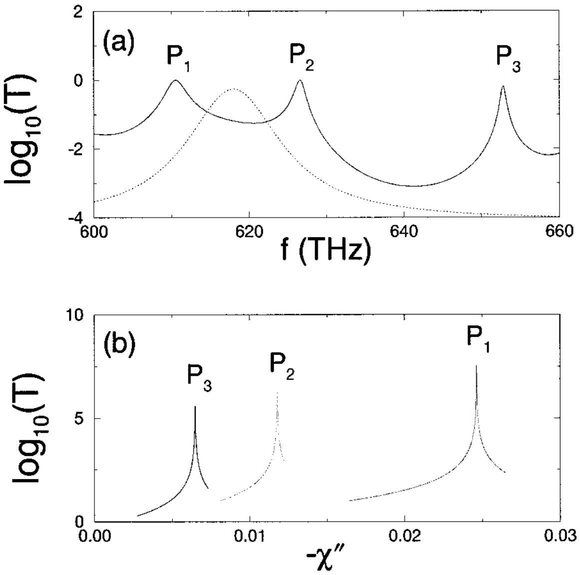

FIG. 1. (a) Logarithm of transmission coefficient vs frequency of a 30-cell random system with three typical peaks. The dotted line shows the frequency dependence of the gain profile. (b) Logarithm of the transmission coefficient vs χ'' for the three peaks shown in (a) with a frequency-independent gain. χ'' is proportional to the pumping rate P_r.

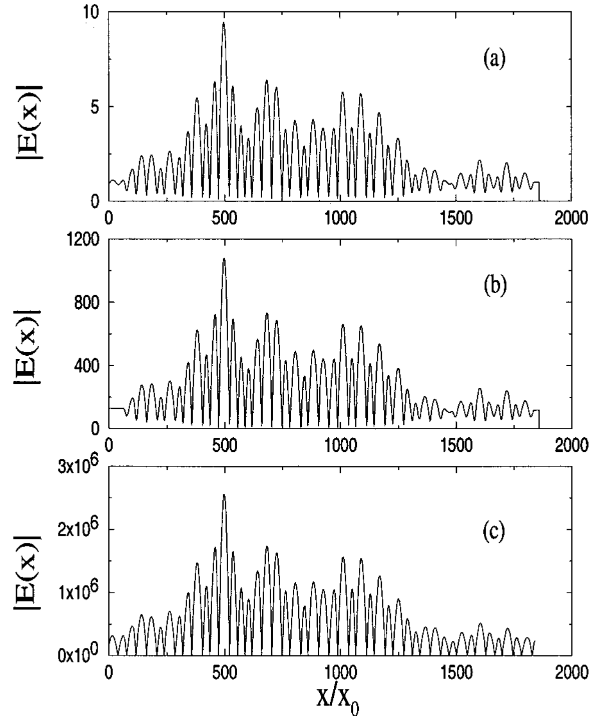

FIG. 2. Amplitude of the electric field of the 30-cell system vs x/x_0, with $x_0 = 5$ nm, for the peak P_2 in Fig. 1a. (a) Without gain, incident field $E_{inc} = 1$ V/m; (b) with near-threshold gain ($P_r = 7.2 \times 10^6$/s), incident field $E_{inc} = 1$ V/m; (c) the lasing field with over-threshold pumping rate ($P_r = 2 \times 10^7$/s) . Both (a) and (b) are obtained by the transfer matrix method while (c) is obtained by the FDTD method and laser theory.

sers can be used to obtain the threshold for lasing. At threshold, the transmission coefficient T goes to infinity. In Fig. 1(b), we plot $\log_{10}(T)$ versus χ'' for the three peaks P_1, P_2, and P_3. In Fig. 1(b), P_3, which has the largest Q, has the smallest threshold for lasing. So, in the frequency-independent gain case, the transfer matrix method indicates that the mode with largest Q will lase first. We have also used the transfer matrix method with a frequency-dependent gain profile, given as a dotted line in Fig. 1(a). We choose the central frequency as $f_a = \omega_a/2\pi = 618.56$ THz, which is exactly between P_1 and P_2, and its width to be $\Delta f_a = \Delta\omega_a/2\pi = 15$ THz. In this case, when we increase the pumping rate P_r, we find that P_2 will lase first and then P_1. So two important conditions determine which mode will lase first; the first is the quality factor of the mode and the second is the gain profile. Experimentally, quite often only part of the random medium has been pumped. Then a third factor comes in, i.e., the spatial overlap of the mode function and the gain region.

The next issue we address is the shape of the wave function, as one introduces gain. In Fig. 2(a), we present the amplitude of the electric field versus distance for $f = 626.6$ THz, which corresponds to the peak P_2 of Fig. 1(a). In Fig. 2(b), we show the wave function with near-threshold gain at the exact high-peak frequency of P_2. Both Figs. 2(a) and 2(b) are obtained by the time-independent transfer matrix method. Notice that in the presence of gain, the shape of the wave function in Fig. 2(b) is almost the same as that without gain in Fig. 2(a). The only change is its amplitude, which increases uniformly. Actually, by keeping the incident amplitude the same, when we increase the gain from zero to the threshold value, we find that the amplitude of the wave function increases from a small to a very large value, but its shape remains almost the same. This is a very interesting result, which is generic since it was also obtained for other localized or "extended" states.

To check if indeed this surprising property is also present is the full time-dependent theory [12,13] of semiclassical laser theories with Maxwell equations, we repeat our calculation for this case too. While in the time-independent theory every mode is independent and amplification is not saturated, this is not true for a real lasing system. In a real lasing system, modes will compete with each other. As was discussed in Ref. [12], in a *short* system, the first lasing mode will suppress all other modes, so we observe only one lasing mode even for a large pumping rate. This is exactly the case when we use the FDTD method to simulate our 30-cell system with an over-threshold gain whose gain profile is the same as given in Fig. 1(a). At first, the electric field is a random one due to spontaneous emission; then a strong lasing mode evolves from the noisy background and a sharp peak appears in the emission spectrum after Fourier transform. The stable field profile of the FDTD calculation is given in Fig. 2(c), where the wave function is the same as in Figs. 2(a) and 2(b), but the amplitude is very large. The emission spectrum of this case is obtained, and gives a sharp peak very close to the resonant peak P_2 of Fig. 1(a). We also checked the wave functions as well as the lasing threshold as we shifted the gain profile. If the central frequency of the gain profile is near P_3, the P_3 mode will lase first, and the shape of the wave function of P_3 remains unchanged. We have also checked the above ideas for a larger ($L \gg \xi$, where ξ is the localization length) system. In such a strong localized case, the form of the wave function remains unchanged as one introduces gain.

The numerical results that are presented above can be explained by the semiclassical Lamb theory [2] of laser physics. According to Lamb theory, the Maxwell equation of the random laser system can be written as

$$-\nabla^2 E(x,t) + \mu_0 \sigma \frac{\partial E(x,t)}{\partial t} + \mu_0 \varepsilon(x) \frac{\partial^2 E(x,t)}{\partial t^2} = -\mu_0 \frac{\partial^2 P_{gain}(x,t)}{\partial t^2}, \tag{1}$$

where μ_0 is the permeability of free space, $E(x,t)$ is the electric field, and the dielectric constant $\varepsilon(x)$ is determined by the random configuration of the system. σ is not just the common conductivity loss, but can be interpreted as the total mode loss including the surface loss of the system by radiation. The polarization due to gain $P_{gain}(x,t)$ [20] is the same as the one defined above.

We assume that the system satisfies the slowly varying approximation (it is always satisfied if we care about the stable lasing state). So $E(x,t) = E_m(t)\phi_m(x)\exp(-i\omega t)$ (our later discussion shows that the separation of the spatial and time parts of the wave function is reasonable), where $E_m(t)$ is the field amplitude, $\phi_m(x)$ is the normalized wave function, and ω is the frequency of the field. The surface loss of the mode is $\sigma_m = \varepsilon_0 \omega / Q_m$, where Q_m is the quality factor of the mode. Thus, we get two equations [2] for the real and the imaginary terms of Eq. (1):

$$\nabla^2 \phi_m(x) + \mu_0[\varepsilon(x) + \varepsilon_0 \chi'(x,\omega)]\omega^2 \phi_m(x) = 0, \tag{2}$$

$$\frac{\partial E_m(t)}{\partial t} = \left(-\overline{\chi''(\omega)} - \frac{1}{Q_m} \right) \frac{\varepsilon_0 \omega E_m(t)}{2\bar{\varepsilon}}, \tag{3}$$

where $\bar{\varepsilon} = \int_0^L \varepsilon(x)dx/L$ is the spatially averaged dielectric constant inside the system, and $\overline{\chi''(\omega)} = \int_0^L \phi_m(x)^* \chi''(x,\omega)\phi_m(x)dx/L$ is the spatially averaged gain. The last integral is done to take into account the overlap between the wave function and the spatial region of gain.

Equation (2) determines the field distribution, quality factor Q_m, and vibration frequency ω of the lasing mode. The term $\varepsilon_0 \chi'(x,\omega)$ will cause the vibration frequency to shift away from the original eigenfrequency of the mode, called the *pulling effect*. For a well-defined mode, generally $Q_m \gg 1$, we need a very small gain to lase. Then $\chi'(x,\omega) \ll 1$, the pulling effect is very weak, and $\omega \simeq \Omega_m$, where Ω_m is the eigenfrequency of the mode. So the wave function of the lasing mode should be similar to the eigenfunction of the mode. Theoretically, we can use the perturbation method to obtain ω and the wave function.

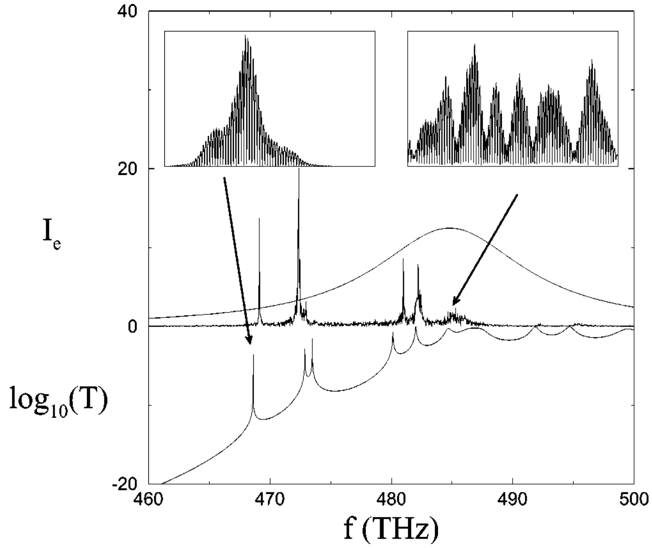

FIG. 3. Logarithm of the transmission coefficient T, the emission intensity I_e, and the gain profile vs frequency f for a 80-cell random systems. In the inset, wave functions of two modes are shown.

Equation (3) is the time-dependent amplitude equation. First we can use it to determine the threshold condition when $-\overline{\chi''}(\omega) = 1/Q_m$. For our system, with homogeneous pumping, we have

$$P_r^c = C_0 \frac{1 + 4(\omega_a - \Omega_m)^2/\Delta\omega_a^2}{Q_m} \tag{4}$$

where $C_0 = [(a_0 + b_0)/a_0](\gamma_c m_e \varepsilon_0 \omega_a \Delta\omega_a / \gamma_r N_0 \tau_{21} e^2)$ is a constant. Equation (4) indeed shows that the threshold value of lasing is inversely proportional to the quality factor Q.

Second, Eq. (3) gives us the stable amplitude of the field when the gain is over threshold. Actually, the gain is saturable, $\chi''(\omega) \propto \Delta N \propto 1/(1 + C_2|E_m|^2)$ [20], in real systems and in our FDTD model [12,13]. With an over-threshold gain, $E_m(t)$ will increase and the gain parameter $\chi''(\omega, E_m)$ will decrease until $-\overline{\chi''}(\omega, E_m) = 1/Q_m$; then the field is stable. So Eq. (3) also determines the amplitude of the stable field for over-threshold pumping cases.

Our numerical and analytical results clearly suggest that states of the random system with gain can easily lase, provided their Q factor is large. These findings provide a path for observing localization of light. Since localized states have large Q values, they will lase with a small pumping rate. On the other hand, strongly fluctuating extended states have smaller Q values because of the radiation loss on the surface of the system and can lase only after a stronger pumping. In a real experiment, even in the presence of absorption, if the gain profile is close to the mobility edge, there is going to be a discontinuity in the critical pumping rate needed for lasing. Localized states will lase first at a low pumping rate, while extended states need a high pumping rate. In Fig. 3, we present the results of for the density of states vs frequency f for a 1d quasidisordered system. On the inset two eigenfunctions are given, one localized at f

$=468$ THz and the other "extended" at $f=485$ THz. In addition the emission intensity I_e vs f is also shown with gain $P_r=10^5$ 1/s, which demonstrates that the localized modes will lase first. In a more realistic 3D case, we expect I_e vs frequency to have many peaks for the localized region and no peaks in the extended region for a given pumping rate [21]. It would be very interesting if this discontinuity can be observed experimentally.

In summary, we have used the time-independent transfer matrix method and the FDTD method to show that all lasing modes come from the eigenstates of the random system. A knowledge of the eigenstates and the density of states of the random system, in conjunction with the frequency profile of the gain, can accurately predict the mode that will lase first, as well as its critical pumping rate. Our detailed numerical results clearly demonstrate that the shape of the wave function remains unchanged as gain is introduced into the system. The role of the gain is to just increase uniformly the amplitude of the wave function without changing its shape. These results can be understood by generalizing the semiclassical Lamb theory of lasing in random systems. These findings can help us to unravel the conditions for observing the localization of light, as well as for manufacturing random lasers with specific emission wavelengths.

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[1] For a recent review, see C.M. Soukoulis and E.N. Economou, Waves Random Media **9**, 255 (1999).
[2] A. Maitland and M.H. Dunn, *Laser Physics* (North-Holland, Amsterdam, 1969), Chap. 9.
[3] Anthony E. Siegman, *Lasers* (University Science Books, Sausalito, CA, 1986), Chaps. 2, 3, 6, and 13.
[4] V.S. Letokhov, Sov. Phys. JETP **26**, 835 (1968).
[5] N.M. Lawandy, R.M. Balachandran, S.S. Gomers, and E. Sauvain, Nature (London) **368**, 436 (1994).
[6] W.L. Sha, C.H. Liu, and R.R. Alfano, Opt. Lett. **19**, 1922 (1994); R.M. Balachandran and N.M. Lawandy, *ibid.* **20**, 1271 (1995); M. Zhang, N. Cue, and K.M. Yoo, *ibid.* **20**, 961 (1995); G. Van Soest, M. Tomita, and A. Lagendijk, *ibid.* **24**, 306 (1999); G. Zacharakis *et al.*, *ibid.* **25**, 923 (2000).
[7] D.S. Wiersma, M.P. van Albada, and Ad Lagendijk, Phys. Rev. Lett. **75**, 1739 (1995); D.S. Wiersma and A. Lagendijk, Phys. Rev. E **54**, 4256 (1996).
[8] H. Cao *et al.*, Phys. Rev. Lett. **82**, 2278 (1999); **84**, 5584 (2000).
[9] S.V. Frolov, Z.V. Vardeny, K. Yoshino, A. Zakhidov, and R.H. Baughman, Phys. Rev. B **59**, R5284 (1999).
[10] S. John and G. Pang, Phys. Rev. A **54**, 3642 (1996), and references therein.
[11] G.A. Berger, M. Kempe, and A.Z. Genack, Phys. Rev. E **56**, 6118 (1997).
[12] Xunya Jiang and C.M. Soukoulis, Phys. Rev. Lett. **85**, 70 (2000).
[13] Xunya Jiang and C.M. Soukoulis, in *Photonic Crystals and Light Localization in the 21st Century*, edited by C.M. Soukoulis (Kluwer, Dordrecht, 2001), p. 417.
[14] H. Cao *et al.* (unpublished).
[15] Xunya Jiang and C.M. Soukoulis, Phys. Rev. B **59**, 6159 (1999).
[16] Xunya Jiang, Qiming Li, and C.M. Soukoulis, Phys. Rev. B **59**, R9007 (1999).
[17] P. Pradhan and N. Kumar, Phys. Rev. B **50**, 9644 (1994); Z.Q. Zhang, *ibid.* **52**, 7960 (1995).
[18] C.W.J. Beenakker Phys. Rev. Lett. **81**, 1829 (1998).
[19] Qiming Li, K.M. Ho, and C.M. Soukoulis, Physica B **296**, 78 (2001).
[20] $\chi'(\omega)=\chi_0''[-\Delta x/(1+\Delta x^2)]$ and $\chi''(\omega)=\chi_0''[1/(1+\Delta x^2)]$, where $\chi_0''=(\gamma_r/\gamma_c)(\Delta N e^2/\varepsilon_0 m_e \omega_a \Delta\omega_a)$ and $\Delta x=2(\omega-\omega_a)/\Delta\omega_a$, $\tau_{21}=1/\gamma_r=1\times10^{-10}$ s is the real lifetime of the upper lasing level, $\gamma_c=(e^2/m)(\omega_a^2/6\pi\varepsilon_0 c^3)$ is the classical decay rate, and $\Delta N=N_2-N_1$ is the number inversion of electrons at the upper and lower lasing levels. Actually $\chi''(\omega,E_m)\propto\Delta N\propto 1/(1+C_2|E_m|^2)$ in our FDTD model and real experimental systems so that the gain is saturable for large field amplitude, but when the field is weak ($<10^5$ V/m), the factor $1/(1+C_2|E_m|^2)$ is very close to 1. ω_a and $\Delta\omega_a$ are the central frequency and linewidth of the gain profile, and e and m_e are the charge and mass of the electron. For a four-level gain medium, $\Delta N=P_r N_0 \tau_{21}$, where $N_0=3.01\times10^{25}$ 1/m^3 is the electron density of the gain medium, and P_r is the pumping rate. With our parameters, we have $P_r=-1.003\times10^{-21}\chi_0''\omega_a\Delta\omega_a$. For details, see [3], Chaps. 2, 3, and 7.
[21] M.N. Skhunor *et al.*, Synth. Met. **116**, 485 (2001), Fig. 7c.