

Dynamic response and relaxation oscillations in random lasers

C. M. Soukoulis,¹ Xunya Jiang,¹ J. Y. Xu,² and H. Cao²

¹Ames Laboratory-USDOE and Department of Physics and Astronomy, Iowa State University, Ames, Iowa 50011

²Department of Physics and Astronomy, Northwestern University, Evanston, Illinois 60208-3112

(Received 27 August 2001; revised manuscript received 2 November 2001; published 2 January 2002)

We experimentally investigate the dynamic response of random lasers using picosecond optical pumping. It is found that the turn-on time of a random laser is less than 100 ps. Above the lasing threshold, the emission pulse is shortened dramatically. The dynamics of each individual lasing mode is different. Lasing starts and gets to a maximum value at different times. In addition, lasing in different modes lasts for different periods of time, and the frequency of some lasing modes also shifts with time. Relaxation oscillations of lasing modes are observed for a random laser. The theoretical simulations based on the semiclassical laser theory and finite different time domain reproduce most of the experimental observations and provide an understanding of the dynamic response of a random laser.

DOI: 10.1103/PhysRevB.65.041103

PACS number(s): 42.25.Bs, 71.55.Jv, 42.55.-f

Both localization and laser theory were developed in the sixties. It was always assumed that disorder was detrimental to lasing action. However, Letokhov¹ theoretically proposed the possibility of lasing in a random system, called random laser. But only after the experimental observation of Lawandy *et al.*,² the random laser systems have been studied intensively.³⁻¹⁰ Since then, many experiments were carried out that showed a drastic spectral narrowing³ and a narrowing of the coherent backscattering peak.⁴ Recently, new experiments^{5,6} showed random laser action with sharp lasing peaks. To fully explain such an unusual behavior of stimulated emission in random systems with gain, a lot of theoretical models were constructed. John and Pang⁷ studied the random lasing system by combining the electron number equations of energy levels with the diffusion equation. Berger *et al.*⁸ have obtained the spectral and spatial evolution of emission from random lasers by using a Monte Carlo simulation. Very recently, Jiang and Soukoulis,^{9,10} by combining a FDTD method with the semiclassical laser theory,^{11,12} were able to study the interplay of localization and amplification. They obtained^{9,10} the field pattern and the spectral peaks of localized lasing modes inside the system. They were able to explain^{9,10} the multi peaks and the nonisotropic properties in the emission spectra seen experimentally.^{5,6} Finally, it was predicted that there is a mode repulsion which gives a saturation in the number of lasing modes in a given random laser system.

Up to now, all the research on random laser is focused on the steady-state properties. The dynamic behavior of random laser modes is rarely studied. For many of the applications of random lasers, the low lasing threshold is very important. However, for other applications, short response time is desired. In this Rapid Communication, we will investigate the dynamic response of random lasers using picosecond optical pumping. It is found that the turn-on time of a random laser is less than 100 ps. Below the lasing threshold, the decay time of the emission pulse hardly decreases with the pump power. Above the lasing threshold, the emission pulse is shortened dramatically. We find that the experimental decay of the emission consists of a fast component due to laser emission and a slow component caused by spontaneous

emission and nonradiative recombination. The dynamics of each individual lasing mode is different. Lasing starts and gets to a maximum value at different times. In addition, lasing in different modes lasts for different periods of time. When the pump power is close to the lasing threshold, relaxation oscillation is observed for the first time in some lasing modes of random lasers. We have also observed that the emission frequency of lasing modes shifts with time. Our theoretical simulations based on the semiclassical laser theory and FDTD reproduce most of the experimental observations and provide an understanding of the dynamic response of a random laser. Our theory is done in 1D, while the experiments are done in 3D. We can obtain qualitative agreement between theory and experiments, because our model contains the correct laser physics concepts (quality factor, gain length, lifetimes of electronic systems, etc.), which are independent of dimensionality.

The sample investigated in our experiments consists of a ZnO powder with an average particle size of 100 nm. The ZnO nanoparticles are deposited onto ITO coated glass substrates by electrophoresis. Thickness of the powder films varies from 10 to 30 μm . The sample is optically excited by the frequency-tripled output ($\lambda = 355$ nm) of a mode-locked Nd:YAG laser (10 Hz repetition rate, 20 ps pulse width). The pump beam is focused to a spot of ~ 20 μm in diameter on the film surface. Emission from the sample is measured by a Hamamatsu streak camera with a 2 ps temporal resolution. The scattered pump light is blocked by the input optics of the streak camera.

Figure 1 shows the temporal evolution of emission below the lasing threshold (a), just above the lasing threshold (b), and well above the lasing threshold (c). Below the lasing threshold, the decay time of the emission is 167 ps. When the pump intensity ($I_e = 80$ MW/cm²) exceeds the lasing threshold, the emission pulse is shortened significantly. As shown in Fig. 1(b), the initial decay of emission is quite fast. The decay time is 27 ps. After ~ 50 ps, the fast decay is replaced by a slow decay. The later decay time is 167 ps, which is equal to the decay time below the lasing threshold. We think that the initial fast decay corresponds to laser emission, and the later slow decay results from spontaneous emis-

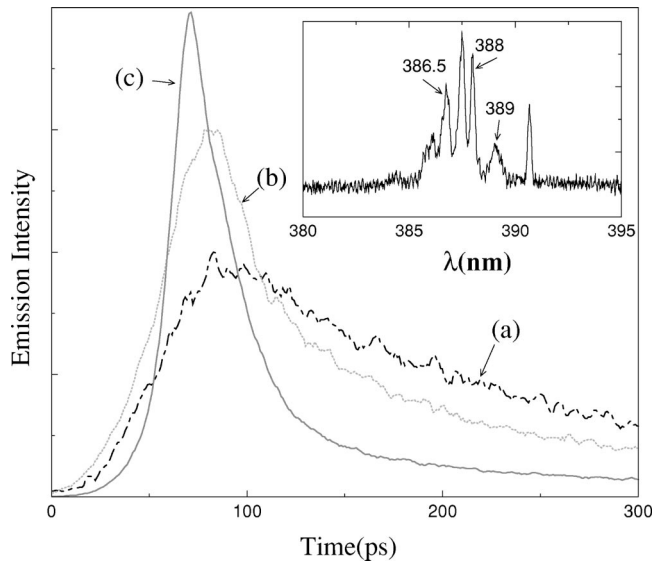


FIG. 1. Temporal evolution of the emission intensity below the lasing threshold (a), just above the lasing threshold (b), and well above the lasing threshold (c). Notice the fast and slow decay in case (c). In the inset the time-integration emission spectrum is shown.

sion and nonradiative recombination. As the pump is increased further, the intensity of the initial laser emission becomes much larger than the intensity of the later spontaneous emission, as shown in Fig. 1(c). As shown in the inset of Fig. 1, there are a few discrete lasing modes in the time-integrated emission spectrum.

Next we study the dynamics of each individual lasing mode (Fig. 2). The emission from the sample is collected by a lens and focused to the entrance slit of a 0.5-m Jarrell-Ash spectrometer. The output port of the spectrometer is connected to the Hamamatsu streak camera whose entrance slit is perpendicular to that of the spectrometer. The photocath-

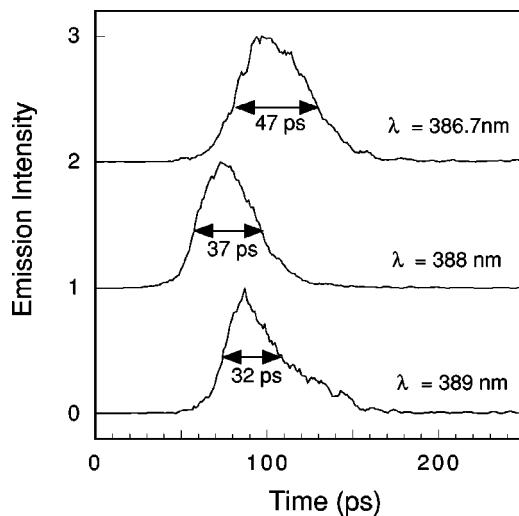


FIG. 2. Temporal profile of the emission profile for three lasing peaks. The mode at 388 nm lases first at 113 ps, while the mode of 389 nm lases secondly at 118 ps, and the 386.7 nm mode lases last at 136 ps.

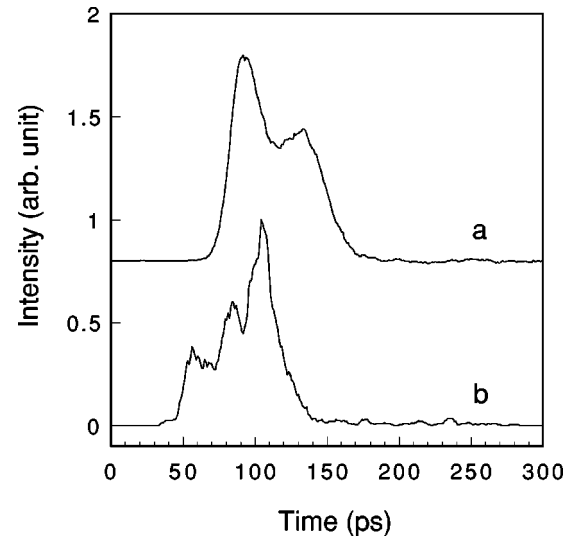


FIG. 3. The temporal evolution of the emission intensity for two different modes above threshold. Relaxation oscillations for random lasers are seen for the first time.

ode width of the streak camera gives an observable spectral window of 6.7 nm with a spectral resolution of 0.1 nm. By combining the spectrometer with the streak camera, we are able to separate different lasing modes and obtain the time trace of each lasing mode. Simultaneously, the time-integrated spectrum of emission from ZnO powder is also measured by a 0.5-meter SPEX spectrometer with a cooled CCD array detector. The temporal profile of three lasing modes are measured by the spectrometer–streak camera apparatus. It's very interesting to notice that the laser emission in these modes is turned on at different times. The mode at 388 nm lases first, and the mode at 386.7 nm starts lasing 23 ps later. In addition, lasing in different modes lasts for different periods of time. The full width at half maximum (FWHM) of laser emission pulses at 386.7 nm, 388 nm, and 389 nm are 47 ps, 37 ps, and 32 ps, respectively. The different temporal behaviors of these three lasing modes suggest that they are not from the same cavity. As we will discuss below, our theoretical model can explain all these interesting observations.

When the pump power is near the lasing threshold, relaxation oscillation is observed for the first time in some of the lasing modes of a random laser.¹⁴ Figure 3 shows the temporal oscillation of emission intensity in two lasing modes. We can see that the oscillation period T is different for these two modes. In Fig. 3(a), $T \approx 23$ ps, and in Fig. 3(b), $T \approx 36$ ps. This again indicates that these two modes are from two cavities with different cavity photon lifetimes. Moreover, the emission intensity increases as it oscillates in Fig. 3(b) while the emission intensity decreases as it oscillates in Fig. 3(a). We find experimentally that for the modes which start lasing earlier, the relaxation oscillation tends to occur in the rising edge of the laser pulse, while for the modes which start lasing later, the relaxation oscillation usually appears at the falling edge of the laser pulse. We speculate that this phenomenon may be caused by the transient optical gain. For instance, for a lasing mode with short turn-on time, the re-

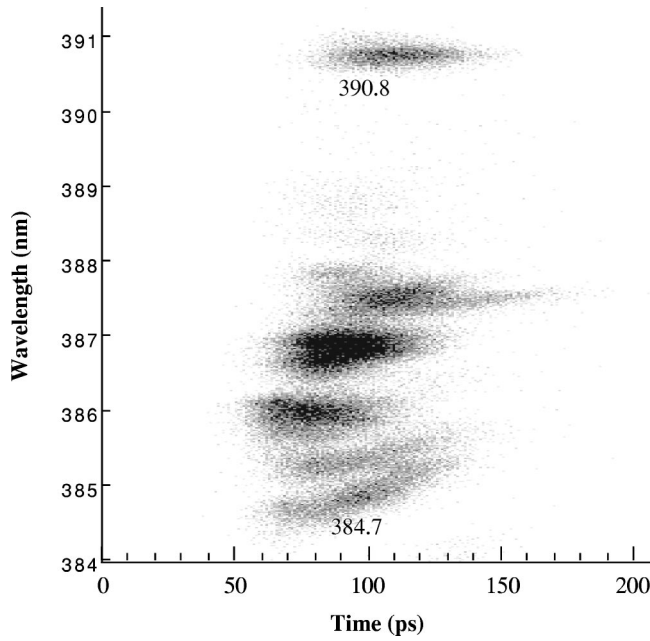


FIG. 4. Time traces of several lasing modes. Both blueshifted and redshifted modes are observed as a function of time.

laxation oscillation occurs while the optical gain still builds up. Thus, the emission intensity increases as it oscillates.

We have also observed how the lasing frequency shifts with time. Figure 4 shows the time traces of several lasing modes when the incident pump pulse energy is 4.5 nJ. The vertical axis corresponds to wavelength, and the horizontal axis represents time. In Fig. 4, the mode at 384.7 nm shifts towards longer wavelengths as time goes on, while the mode

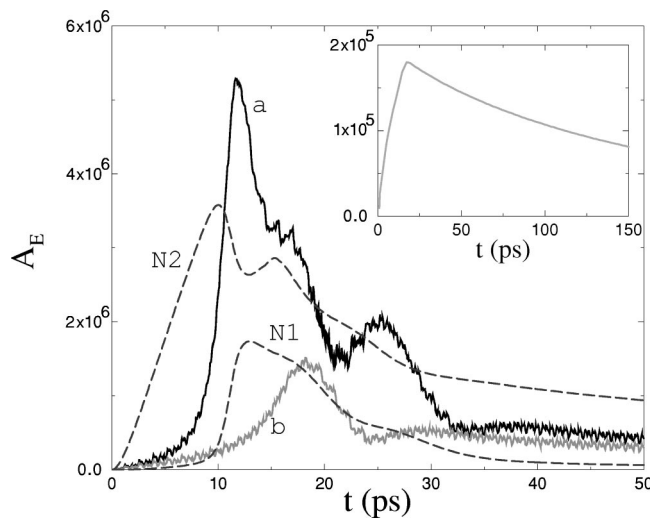


FIG. 5. The amplitude of the electric field A_E at the center point of a mode vs t for different pumping intensities, line (a) for a pumping intensity just above threshold, line (b) for a well above threshold pumping intensity, the inset is for an under-threshold pumping case. The dashed lines show the electron populations on two lasing levels at the center point of the mode vs t for the case (b). The higher dashed line is the population of the (N_2) lasing level, while the lower is for the (N_1) level.

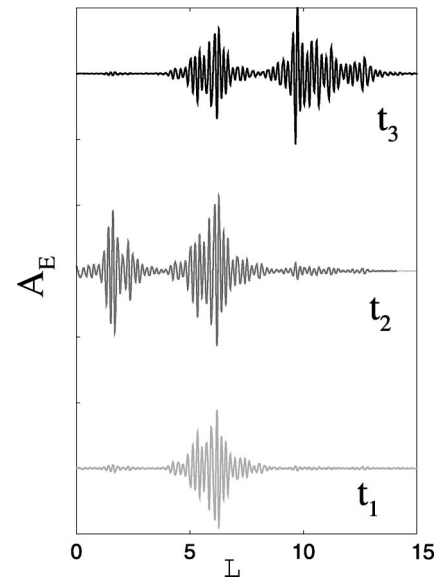


FIG. 6. The amplitude of the electric field A_E vs the position L of the system, for three times of observation. At $t_1=13$ ps, only one mode lases, at $t_2=17.5$ ps, both modes are lasing, while for $t_3=24.7$ ps, the central mode is still lasing, and a new mode starts lasing.

at 390.8 nm remains nearly constant in frequency as time goes on. We have repeated the same measurements at different positions of the sample. We find that the frequency shift varies in random manner from mode to mode. There are modes that are blue-shifted with time, while others are red-shifted. Finally, we have observed sudden frequency jump of some lasing modes. A mode lases for some time at a given frequency and then jumps suddenly to another frequency.

We have introduced a model^{9,10} for the dynamic response and relaxation oscillation in random lasers, the results of which are tested by a comparison with the experiments presented above. The formalism is dynamical and can treat the

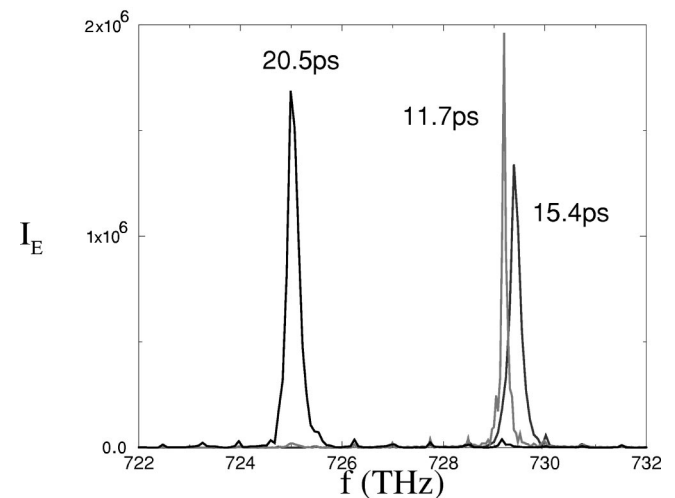


FIG. 7. The emission spectra of the three modes shown in Fig. 6 vs the frequency for different times of observation. Notice that the peak at the center lases first at 11.7 ps, then the right peak at 15.4 ps, and last the left peak at 20.5 ps.

random laser system above threshold too. A random laser is intrinsically time-dependent due to the very different time scale of transport and population dynamics. In Fig. 5, we present the theoretical results for the temporal evolution of the amplitude of the electric field A_E below the lasing threshold in the insert of Fig. 5, just above threshold (a), and well above the lasing threshold (b). Notice that there is a qualitative agreement between the theoretical results presented in Fig. 5, with the experimental results presented in Fig. 1. Below threshold from the insert of Fig. 5 we obtain the decay time is $\tau=112$ ps, at threshold [Fig. 5(a)] the fast decay time is 4 ps, while the longer decay time is again 112 ps. From Fig. 5(b), we can see that relaxation oscillation are also observed in our simulation. The period $T\approx 5$ ps, which is much smaller than the experimental one ($T\approx 36$ ps) seen in Fig. 3(b). This discrepancy is due to the fact that our model is a 1D system in which the modes have higher quality factors Q than the 3D experimental modes. In Fig. 5, also shown the N_2 and N_1 vs t . Notice that N_2 drops substantially as N_1 still increases when the field increase fast. At the same time the field gets to its maximum value. So the local dynamic processes of field can influence the local gain in the random material and this is why this model can physically explain most dynamic phenomena.

Our numerical simulations can follow the evolution of A_E as we increase the pumping rate. In Fig. 6, we present spatial evolution of A_E for different times of observation. In all the cases shown in Fig. 6, the pumping time is 15 ps. Notice that for $t_1=13$ ps, A_E is strongly localized at one position, as the

observing time t increases further ($t_2=17.5$ ps), two localized modes are seen. For even longer observing time ($t_3=24.7$ ps), a new localized state emerges. If we take the Fourier transform of the time evolution of the mode in Fig. 6, we find that the emission intensity gives a peak at $f=729.2$ THz. For the second and third lasing modes (in Fig. 7) we obtain the frequency peaks at 729.4 THz and 725.0 THz, respectively. These results are in qualitative agreement with those presented in Fig. 2. It is clear that the different temporal behavior of these three modes suggests that they are coming from different localized cavities.

In summary, we have presented an experimental and theoretical study of the dynamic response and relaxation oscillations of random lasers. It is found that the turn-on time of a random laser is less than 100 ps. Above the lasing threshold, the emission pulse is shortened dramatically and relaxation oscillations are observed. The full dynamic of the interplay between the field and the electron populations is found to be an essential part of a description of a random laser close to or above the lasing threshold. In further studies, it might be possible to tailor a random laser system to absorb in one particular wavelength and emit in different wavelengths. The random laser turn-on time can be tailored at will.

Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. W-7405-Eng-82. This work was supported by the director for Energy Research, Office of Basic Energy Sciences.

¹V.S. Letokhov, Sov. Phys. JETP **26**, 835 (1968).

²N.M. Lawandy, R.M. Balachandran, S.S. Gomers, and E. Sauvain, Nature (London) **368**, 436 (1994).

³W.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).

⁴D.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).

⁵H. Cao *et al.*, Phys. Rev. Lett. **82**, 2278 (1999); H. Cao *et al.*, *ibid.* **84**, 5584 (2000).

⁶S.V. Frolov, Z.V. Vardeny, K. Yoshino, A. Zakhidov, and R.H. Baughman, Phys. Rev. B **59**, R5284 (1999).

⁷S. John and G. Pang, Phys. Rev. A **54**, 3642 (1996), and references therein.

⁸G.A. Berger, M. Kempe, and A.Z. Genack, Phys. Rev. E **56**, 6118 (1997).

⁹Xunya Jiang and C.M. Soukoulis, Phys. Rev. Lett. **85**, 70 (2000).

¹⁰Xunya 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.

¹¹C.W.J. Beenakker, Phys. Rev. Lett. **81**, 1829 (1998).

¹²A. Maitland and M. H. Dunn, *Laser Physics* (North-Holland, Amsterdam, 1969), Chap. 9.

¹³Anthony E. Siegman, *Lasers* (Mill Valley, California, 1986), Chaps. 2, 3, 6, and 13.

¹⁴See, for example, an experimental observation of multipulsing of emission by C. Gouedard *et al.*, J. Opt. Soc. Am. B **10**, 2358 (1993).