Transition from amplified spontaneous emission to laser action in strongly scattering media

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In an active random medium, the combination of multiple scattering with light amplification may lead to random laser action. However, it is crucial but sometimes difficult to distinguish between amplified spontaneous emission and lasing. By varying the amount of scattering in an amplifying random medium, we have observed the transition from amplified spontaneous emission to lasing with coherent feedback. We have found out when the transition occurs through the measurement of the scattering mean free path. Our numerical simulation based on the direct solution to Maxwell equations clearly illustrates the transition from light amplification to laser oscillation due to an increase of the amount of scattering in active random medium.

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Since the first observation of laserlike emission from laser dye solution containing microparticles [1], there has been considerable debate on whether it is amplified spontaneous emission (ASE) or true laser emission [2]. Although a dramatic spectral narrowing occurs above a pump threshold, discrete lasing modes are missing [1,3]. Since the scattering mean free path l is much longer than the emission wavelength λ ($l \gg \lambda$), scattering merely increases the path length of light in the gain region, but cannot provide coherent feedback [4]. Thus, what has happened is amplification of spontaneous emission, or loosely speaking, lasing with nonresonant feedback [5]. The experimental results can be explained by the model of light diffusion with gain, where the phase of light wave and interference effect are neglected [6–8].

Recently, we have observed random laser action with resonant feedback in semiconductor powder [9]. Since the scattering mean free path l is close to the emission wavelength λ ($l \sim \lambda$), recurrent light scattering events arise and provide coherent feedback for lasing. Above a pump threshold, discrete lasing modes appear in the emission spectrum in addition to a drastic increase of emission intensity. The dependence of the lasing threshold pump intensity on the excitation volume agrees with that predicted by the random laser theory [10].

Therefore, the behavior of an active random medium in the regime of incipient photon localization ($l \sim \lambda$) is very different from that in the diffusion regime ($l \gg \lambda$). The questions we will address here are (1) when the transition from amplified spontaneous emission to lasing with coherent feedback occurs; (2) whether the transition is a gradual or sudden transition.

To study this transition, we use laser dye solution containing semiconductor nanoparticles. The advantage of the suspension is that the gain medium and the scattering elements are separated. Thus, we can independently vary the amount of scattering by particle density and the optical gain by dye concentration.

Experimentally, rhodamine 640 perchlorate dye and zinc oxide (ZnO) particles are mixed in methanol. The ZnO particles have a mean diameter of 100 nm. To keep the particles from clustering, the solution, contained in a flask, is shaken in an ultrasonic cleaner for 20 min right before the photoluminescence experiment. The frequency-doubled output ($\lambda = 532$ nm) of a mode-locked Nd:YAG laser (10 Hz repetition rate, 25 ps pulse width) is used as pump light. The pump beam is focused by a lens (10 cm focal length) onto the solution contained in a 1 cm$\times$1 cm$\times$3 cm cuvette at nearly normal incidence. Emission into the direction $\sim 45°$ from the normal of the cell front window is collected by a fiber bundle and directed to a 0.5-m spectrometer with a cooled CCD detector array.

By changing the ZnO particle density in the solution, we continuously vary the amount of scattering in order to map out the transition from ASE to lasing. Figures 1 and 2 show the evolution of the emission spectra with the pump intensity when the ZnO particle density is $\sim 2.5\times 10^{11}$ cm^{-3} and $\sim 1.0\times 10^{12}$ cm^{-3}, respectively. The dye concentration is fixed at 5×10^{-3} M. As shown in the inset of Fig. 1, when the incident pump pulse energy exceeds ~ 3 μJ, the emission linewidth is quickly reduced to ~ 5 nm, meanwhile the peak intensity increases much more rapidly with the pump power. This is because optical scattering by the ZnO particles increases the path length of the emitted light inside the gain region. When the photon travels in the gain regime, it may induce the stimulated emission of a second photon. As the pump power increases, the gain length is reduced. Eventually the gain length at frequencies near the maximum of the gain spectrum approaches the average path length of the photons in the gain regime. Then the probability of a photon generating a second photon before leaving the gain regime approaches one, and the emission intensity suddenly increases. From the theoretical point of view, the solution to the diffusion equation including optical gain diverges. The drastic increase of the emission intensity at frequencies near the maximum of the gain spectrum results in a significant narrowing of the emission spectrum. This phenomenon is similar to the neutron scattering in combination of nuclear fission [11].

However, when the ZnO particle density increases, the

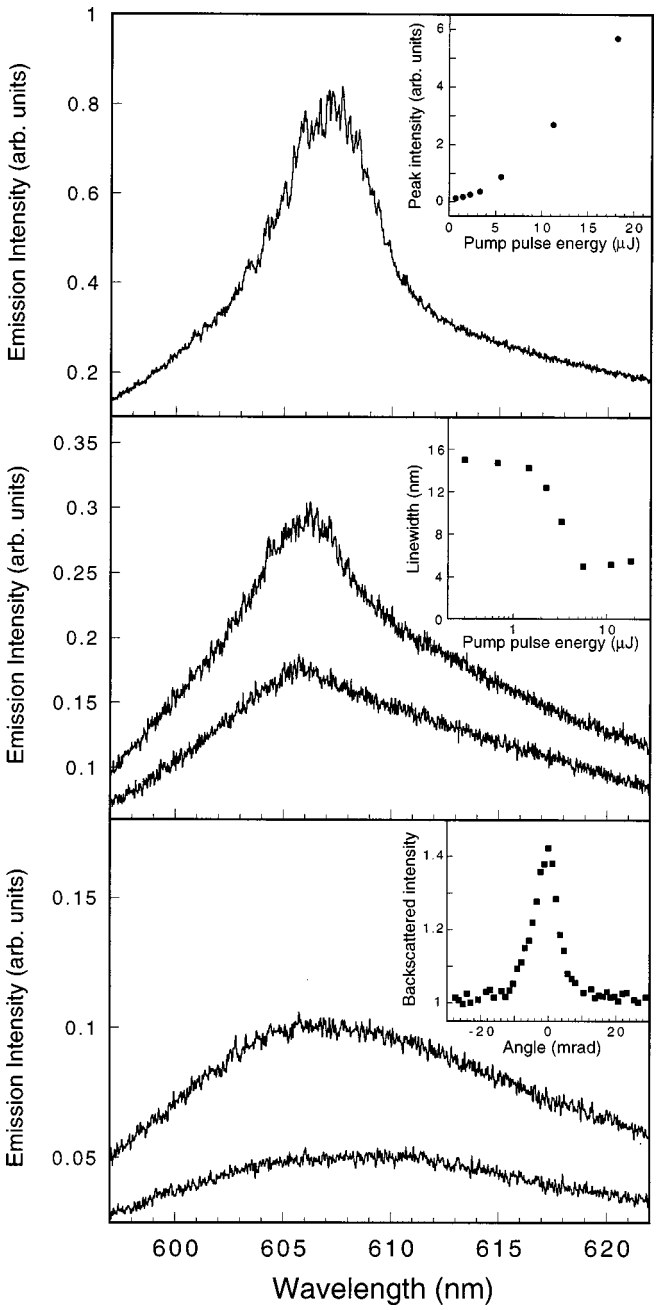

FIG. 1. Emission spectra when the incident pump pulse energy is (from bottom to top) 0.68, 1.5, 2.3, 3.3, 5.6 μJ. The ZnO particle density is $\sim 3\times 10^{11}$ cm^{-3}. The upper inset is the emission intensity at the peak wavelength versus the pump pulse energy. The middle inset is the emission linewidth versus the pump pulse energy. The lower inset is the coherent backscattering cone.

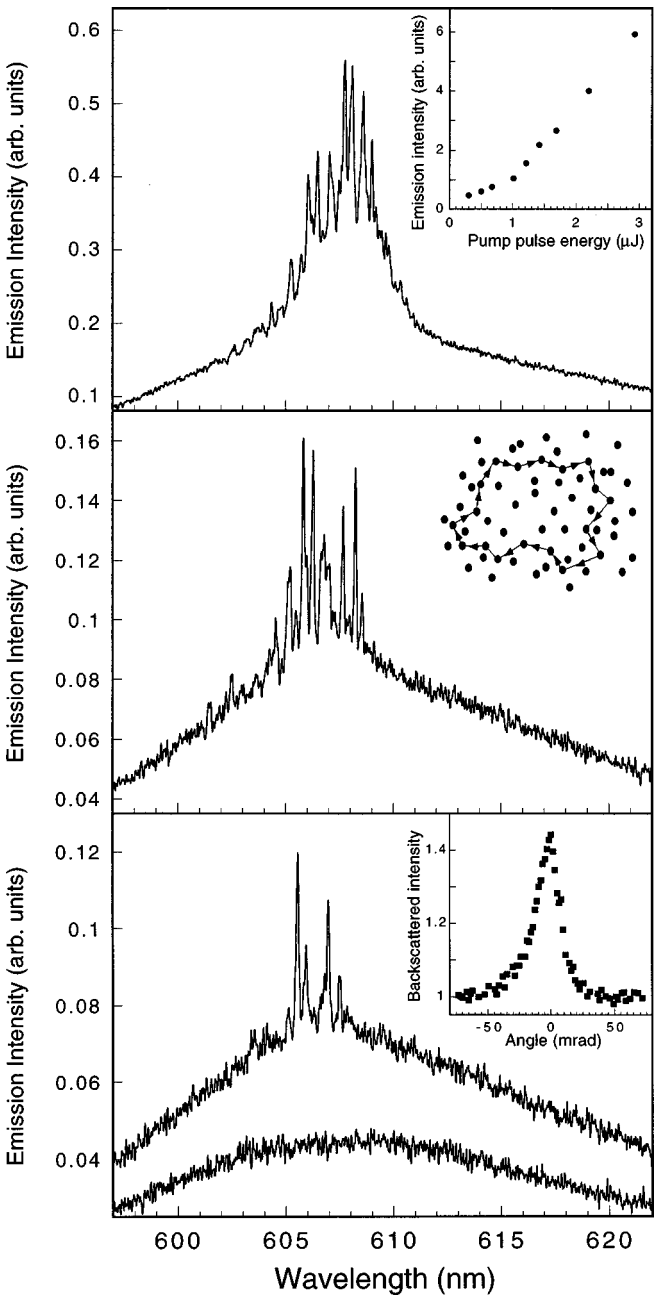

FIG. 2. Emission spectra when the incident pump pulse energy is (from bottom to top) 0.68, 1.1, 1.3, 2.9 μJ. The ZnO particle density is $\sim 1\times 10^{12}$ cm^{-3}. The upper inset shows the emission intensity integrated over a regime near the peak wavelength versus the pump pulse energy. The middle inset is a schematic diagram showing the formation of a closed loop path for light through recurrent scattering in a random medium. The lower inset is the coherent backscattering cone.

phenomenon becomes dramatically different. As shown in Fig. 2, when the incident pump pulse energy exceeds 1.0 μJ, discrete peaks emerge in the emission spectrum. The linewidth of these peaks is less than 0.2 nm, which is more than 50 times smaller than the ASE linewidth below the threshold. When the pump intensity increases further, more sharp peaks appear. These discrete peaks result from recurrent light scattering. As shown schematically in the inset of Fig. 2, when the ZnO particle density is high enough, the emitted light may return to a scatterer from which it is scattered before, and thereby forming a closed loop path. When the amplification along such a loop path exceeds the loss, laser oscillation can occur in the loop which serves as a laser resonator. The requirement of the phase shift along the loop being equal to a multiple of 2π determines the oscillation frequencies. Laser emission from these cavities results in discrete narrow peaks in the emission spectrum. Because the ZnO particles are mobile in the solution, the frequencies of the lasing modes changes from pulse to pulse. The emission spectra in Figs. 1 and 2 are taken for a single pump pulse. When the pump power increases further, the gain exceeds the

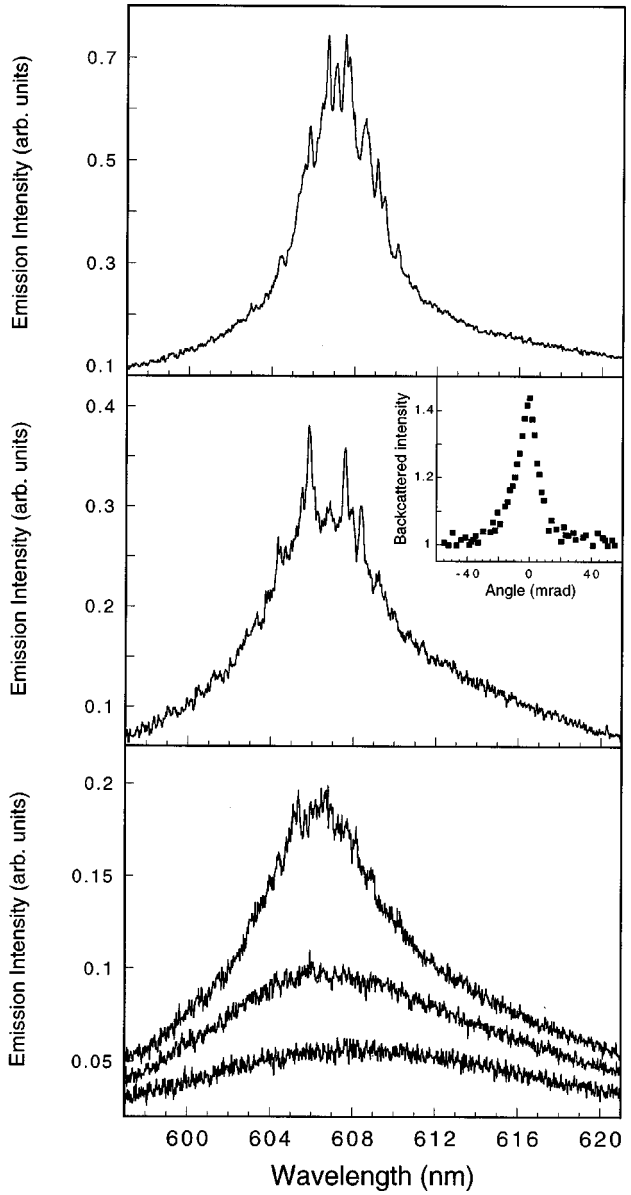

FIG. 3. Emission spectra when the incident pump pulse energy is (from bottom to top) 0.74, 1.35, 1.7, 2.25, and 3.4 μJ. The ZnO particle density is $\sim 6\times 10^{11}$ cm^{-3}. The inset is the coherent backscattering cone.

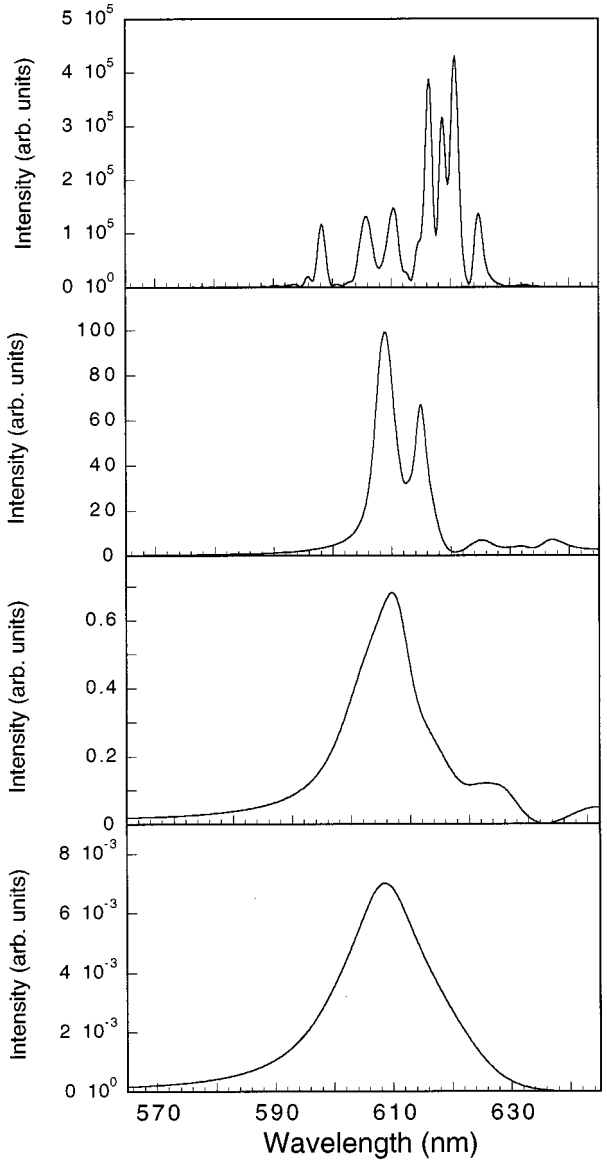

FIG. 4. Calculated emission spectra when the ZnO particle densities in the dye solution are (a) 2.3×10^{7} cm^{-2}, (b) 1.1×10^{8} cm^{-2}, (c) 2.3×10^{8} cm^{-2}, and (d) 2.3×10^{9} cm^{-2}.

loss in more cavities formed by multiple scattering. Laser oscillation in those cavities results in more discrete peaks in the emission spectrum.

To find out when the transition from ASE to lasing occurs, we have measured the scattering mean free path l in the methanol solution of ZnO particles in the coherent backscattering (CBS) experiment [12,13]. The output from a He:Ne laser is used as the probe light since its wavelength is close to the emission wavelength of rhodamine 640 perchlorate dye. The measured backscattering cones from the solutions are shown in the lower inset of Figs. 1 and 2. From the angle of cusp, we estimate that $l\approx 14\lambda$ in Fig 1, and $l\approx 5\lambda$ in Fig. 2. Thus the transition from ASE to lasing occurs when the scattering mean free path is decreased from 14λ to 5λ.

Furthermore, we find the transition from amplified spontaneous emission to lasing with coherent feedback is a gradual one. Figure 3 shows the evolution of the emission spectra with the pump intensity when the ZnO particle density is $\sim 5\times 10^{11}$ cm^{-3}. From the coherent backscattering cone shown in the inset of Fig. 3, we estimate the scattering mean free path $l\simeq 8\lambda$. As the pump power increases, a drastic spectral narrowing occurs first. Then at higher pump intensity, discrete narrow peaks emerge in the emission spectrum. In the solution, there is some but not large probability of a photon scattered back to the same scatterer from which it is scattered before. In other words, the cavities formed by multiple scattering is quite lossy. The pump intensity required to reach lasing threshold in these cavities is high. Thus, the pump intensity first reached the threshold where the gain length near the maximum of the gain spectrum becomes equal to the average path length of photons in the excitation volume. A significant spectral narrowing and a sudden increase of peak emission intensity occur, similar to what happens in Fig. 1. Then the pump intensity reaches a second threshold where the amplification along some closed loop paths formed by scattering exceeds the loss. Lasing os-

cillation occurs in these cavities, adding discrete peaks to the emission spectrum. However, the number of lasing modes in Fig. 3 is less than that in Fig. 2 under similar pump power. When the gain length and excitation volume are the same, a longer scattering mean free path results in less number of loops where the lasing threshold can be reached. As the ZnO particle density is increased, the lasing threshold density decreases.

Several models have been set up in the theoretical study of stimulated emission in active random media, e.g., diffusion equation with gain [4,6], Monte Carlo simulation [8], ring laser with nonresonant feedback [7]. However, these models cannot predict lasing with coherent feedback because the phase of the optical field is neglected. We take a different approach: namely, we directly calculate the electromagnetic field distribution in the random medium by solving the Maxwell equations using the finite-difference time-domain (FDTD) method [14]. However, the simulation of three-dimensional random media using the FDTD method requires much computing power. Here we present our simulation of the transition from ASE to lasing in two-dimensional random media.

In our model, ZnO particles, either round or square in shape, randomly distributed in dye solution. The particle size is 70 nm. The excitation area is 10×10 μm. We use the uniaxial perfect matched layer (UPML) absorbing boundary condition [15]. For TE mode, the Maxwell curl equations are

$$\frac{\partial H_x}{\partial t}=-\frac{1}{\mu_0}\frac{\partial E_z}{\partial y},$$

$$\frac{\partial H_y}{\partial t}=\frac{1}{\mu_0}\frac{\partial E_z}{\partial x}, \tag{1}$$

$$J_z+\epsilon\frac{\partial E_z}{\partial t}=-\frac{\partial H_x}{\partial y}+\frac{\partial H_y}{\partial x}.$$

We introduce optical gain by negative conductance [16]. The spectral gain profile of the dye solution is

$$\sigma(\omega)\equiv\frac{J_z(\omega)}{E_z(\omega)}=-\frac{\sigma_0}{2}\left(\frac{1}{1+i(\omega-\omega_0)T_2}+\frac{1}{1+i(\omega+\omega_0)T_2}\right). \tag{2}$$

σ_0 is related to the peak value of the gain set by the pumping level. T_2 is the dipole relaxation time, which is inversely proportional to the spectral gain width. Within a semiclassical framework, spontaneous emission can be included in Maxwell's equation as a noise current [17]. By solving Eq. (1) using the FDTD method, we obtain the electrical field $E_z(t)$. Using the discrete Fourier transformation of $E_z(t)$, we obtain the emission spectrum $I(\omega)=|E(\omega)^2|$.

In our simulation, we fix the optical gain and vary the particle density. Figure 4 shows the calculated emission spectra at different particle densities. The parameters used in this calculation are $\sigma_0=5\times10^4$ Ohm/m, $T_2=1.3\times10^{-14}$ s, $\lambda_0=605$ nm. At the particle density of 2.3×10^7 cm^{-2}, the emission spectrum is quite broad. When the particle density is increased to 1.1×10^8 cm^{-2}, the emission spectrum is narrowed. This is because the scattering by ZnO particles increases the path length of photons in the active random medium. The emitted light whose frequency near the center of the gain spectrum is amplified preferentially. When the particle density becomes 2.3×10^8 cm^{-2}, a couple of discrete peaks emerge in the emission spectrum. These peaks indicate the formation of optical cavities by multiple scattering. Eventually when the particle density is increased to 2.3×10^9 cm^{-2}, more discrete peaks emerge, and the spectral linewidth of the discrete peaks is reduced. Due to the increase of the amount of scattering, more cavities are formed by light scattering, and the cavity quality factor is improved.

In addition, we have calculated the temporal evolution of the total emission intensity. At the particle density of 2.3×10^9 cm^{-2}, the emission intensity increases with time. However, when the particle densities are 2.3×10^7 cm^{-2}, 1.1×10^8 cm^{-2}, and 2.3×10^8 cm^{-2}, the emission intensity does not increase with time. Hence, lasing oscillation occurs at the particle density of 2.3×10^9 cm^{-2}, while there is only amplified spontaneous emission in the other three cases of lower particle densities. Our simulation result demonstrates that an increase of particle density leads to the transition from ASE to lasing in the active random medium.

In summary, by continuously varying the amount of scattering in an active random medium, we have observed the transition from amplified spontaneous emission to lasing with coherent feedback. In the strong scattering regime, recurrent light scattering events arise and provide resonant feedback for lasing. Our numerical simulation based on the direct solution to Maxwell equations illustrates the transition from light amplification to laser oscillation due to an increase of the amount of scattering in active random medium.

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