Random Laser Action in Semiconductor Powder

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We report the first observation of random laser action with coherent feedback in semiconductor powder. Since the scattering mean free path is less than the emission wavelength, recurrent light scattering arises and provides coherent feedback for lasing. Discrete lasing modes have been observed above the threshold. The dependence of the lasing threshold intensity on the excitation volume agrees with the random laser theory. Laser emission from the powder could be observed in all directions. This observation also provides direct evidence for the existence of recurrent scattering of light. [S0031-9007(99)08739-6]

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Anderson localization of electrons in a disordered system has become a prominent part of contemporary condensed matter physics, and remains an active subject of theoretical and experimental research [1]. Since Anderson localization is completely based on the interference effect, and interference is a common property of all wave phenomena, it is natural to extend electron localization to photon localization in disordered dielectric media. Recently, Anderson localization of photons was observed in GaAs powder [2].

Apart from the remarkable similarities, there are striking differences between electron transport and photon transport in a disordered medium. For example, the number of electrons is always conserved, while the number of photons is not in an amplifying (or absorbing) random medium. A fascinating phenomenon, which would never occur in an electronic system, is laser action in a disordered gain medium. In the case of strong scattering and gain, recurrent scattering events could provide coherent feedback and results in lasing [3]. As shown in the inset of Fig. 2, when the scattering mean free path becomes equal to or less than the wavelength, light may return to a scatterer from which it was scattered before, and thereby forming closed loop paths. If the amplification along such a loop path exceeds the loss, laser oscillation could 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. Such a laser is called a "random laser." It represents a coherent effect in an active random medium.

There has been tremendous interest in and search for random lasers over the past few years. Laserlike emission has been observed from laser dye solutions containing microparticles and from laser crystal powders [4–8]. Although a drastic spectral narrowing has been observed, discrete lasing modes were missing. This is because the scattering mean free path is much longer than the optical wavelength. Scattering merely increases the path length of light in the gain region, but can not provide coherent feedback which is essential to laser action. Thus, what was observed is amplified spontaneous emission rather than true lasing. The experimental observation could be well explained by the model of light diffusion with gain, where the phase of light wave and interference effect are neglected [9–11].

The main difficulty to realize a random laser has been the simultaneous realization of strong scattering and high gain. We have been able to realize such conditions in zinc oxide (ZnO) and gallium nitride (GaN) powder, and observed random laser action.

ZnO powder with an average particle size of 100 nm was deposited onto ITO coated glass substrates by electrophoresis [12]. Thickness of the powder films varied from 6 to 15 μm.

We characterized the scattering mean free path l in the coherent backscattering (CBS) experiment [13–15]. ZnO has a direct band gap of 3.3 eV. To avoid absorption, the frequency-doubled output ($\lambda = 410$ nm) of a mode-locked Ti:sapphire laser (76 MHz repetition rate, 200 fs pulse width) was used as the probe light. Figure 1 shows the measured backscattering cone of the ZnO powder film. From the angle of cusp, we estimated that $l \approx 0.8\lambda$, after taking into account internal reflection [16–18]. Since the wavelength (380–390 nm) of ZnO emission is quite close

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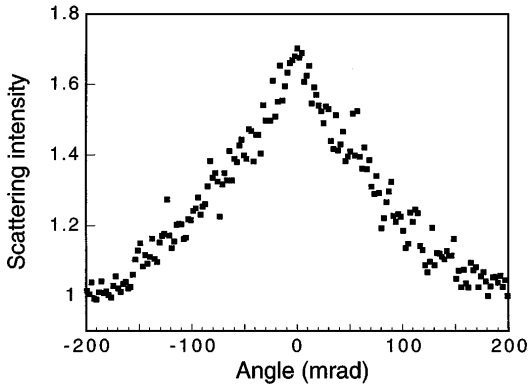

FIG. 1. Coherent backscattering cone from a ZnO powder film. The film thickness is 15 μm.

to the probe wavelength, the scattering mean free path of the emitted light is close to that of the probe light. To directly obtain the scattering mean free path at the emission wavelength, one needs to numerically simulate CBS taking into account the absorption and compare it with the experimental data.

In the photoluminescence experiment, the samples were optically pumped by the frequency-tripled output (λ = 355 nm) of a mode-locked Nd:YAG laser (10 Hz repetition rate, 15 ps pulse width). The pump beam was focused to a spot or a stripe on the film surface with normal incidence. Electrons in the valence band absorbed pump photons and jumped to the conduction band. They subsequently relaxed to the bottom of the conduction band before radiative decay. By comparing the spontaneous emission intensity from a 6 μm thick ZnO powder film and that from ZnO single crystal, we estimated that only $\sim$1% of the pump light was absorbed by the ZnO powder, and the rest was scattered. Figure 2 shows the evolution of the emission spectra as the pump intensity was increased. At low excitation intensity, the spectrum consisted of a single broad spontaneous emission peak. As the pump power increased, the emission peak became narrower due to the preferential amplification at frequencies close to the maximum of the gain spectrum. When the excitation intensity exceeded a threshold, very narrow peaks emerged in the emission spectra. The linewidth of these peaks was less than 0.3 nm, which was more than 30 times smaller than the linewidth of the amplified spontaneous emission peak below the threshold. When the pump intensity increased further, more sharp peaks appeared. The frequencies of the sharp peaks depends on the sample position. In other words, when we moved the excitation spot across the film, the frequencies of the sharp peaks changed. Figure 3 shows the spectrally integrated emission intensity as a function of the excitation intensity. A threshold behavior was observed: above the pump power at which multiple sharp peaks emerged in the emission spectrum, the integrated emission intensity increased much more rapidly with the pump power. These

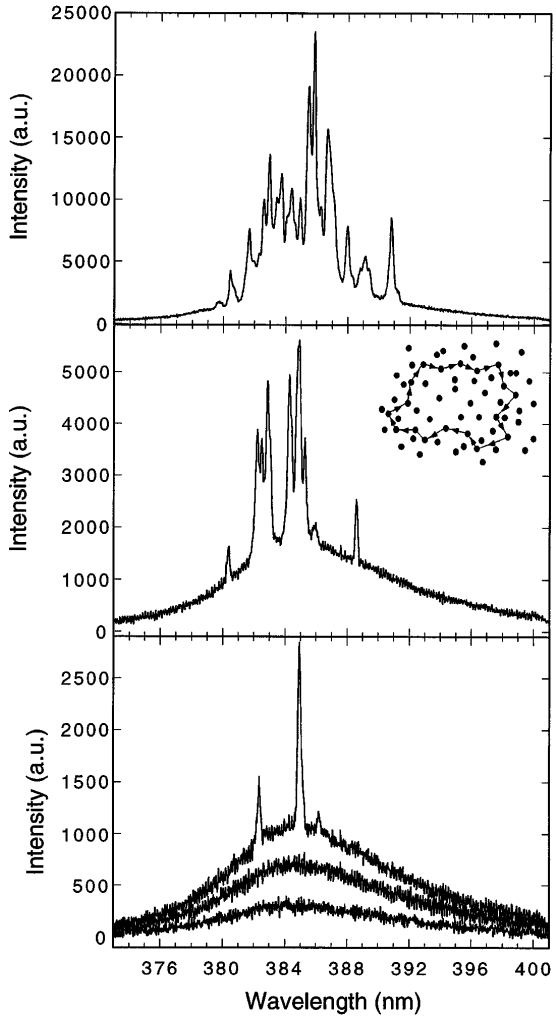

FIG. 2. Spectra of emission from ZnO powder when the excitation intensity is (from bottom to top) 400, 562, 763, 875, and 1387 kW/cm^2. The thickness of the film of ZnO powder is 6 μm. The excitation area is about 1600 μm^2. The inset is a schematic diagram showing the formation of a closed loop path for light through recurrent scattering in the powder.

data suggest that laser action has occurred in the ZnO powder.

Because of the short scattering mean free path in the ZnO powder, the emitted light is strongly scattered, and closed loop paths can be formed through multiple scattering. There are many such closed loop paths in the powder. These loops could serve as ring cavities for light. Along different loop paths, the probability of a photon scattered back to its starting point is different. In other words, the ring cavities formed by recurrent scattering have different loss. When the pump intensity increases, the gain reaches the loss first in the low-loss cavities. Then laser oscillation occurs in these cavities, and the lasing frequencies are determined by the cavity resonances. Laser emission from these resonators results in a small number of discrete narrow peaks in the emission spectrum. As the pump power increases further, the gain increases and it exceeds the loss

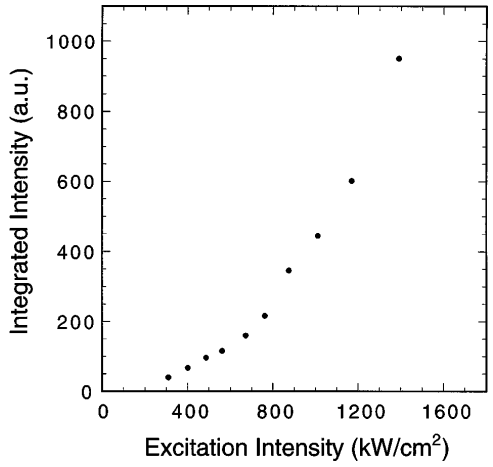

FIG. 3. Spectrally integrated intensity of emission from the ZnO powder versus the excitation intensity.

in the lossier cavities. Laser oscillation in those cavities adds more discrete peaks to the emission spectrum.

For an amplifying random medium, the relevant length scales are the scattering mean free path l, the gain length l_g, and the sample size. In Fig. 2, the excitation volume $V = 9600\ \mu\text{m}^3$. According to the random laser theory [3,19], the gain length at the onset of lasing is $l_g \sim V^{2/3}/l \sim 2.8$ mm. Experimentally, at the lasing threshold pump intensity (763 kW/cm^2), the gain coefficient was measured to be $\sim$20 cm^{-1}. The corresponding gain length is $\sim$1 mm. Hence, the measured lasing threshold intensity is in reasonable agreement with that predicted by the random laser theory.

We have studied the dependence of random laser action on the excitation volume. Figure 4 shows the evolution of the emission spectra as we changed the excitation area while keeping the pump intensity constant. When the excitation area was increased, more lasing peaks emerged in the emission spectra. Since there are more closed loop paths for light in a larger excitation volume, laser action could occur in more cavities formed by recurrent scattering. On the other hand, when the excitation area was reduced to below a critical size, laser oscillation stopped. Since the closed loop paths were too short, the amplification along the loops was not high enough to achieve lasing.

Random laser theory predicts a critical sample size for lasing [3,19]. For a three-dimensional random medium, the critical volume $V \sim (ll_g)^{3/2}$. In Fig. 4, the gain length $l_g \sim 0.75$ mm, and thus the critical volume $V \sim 1290\ \mu\text{m}^3$. Experimentally, the critical volume $V = 5940\ \mu\text{m}^3$. The measured critical volume is 4.6 times larger than the theoretical estimation. One reason for the discrepancy is that the geometrical factor is neglected in the estimation of the critical volume.

Unlike a conventional laser, laser emission from the semiconductor powder could be observed in all directions. As shown in Fig. 5, the laser emission spectra varied

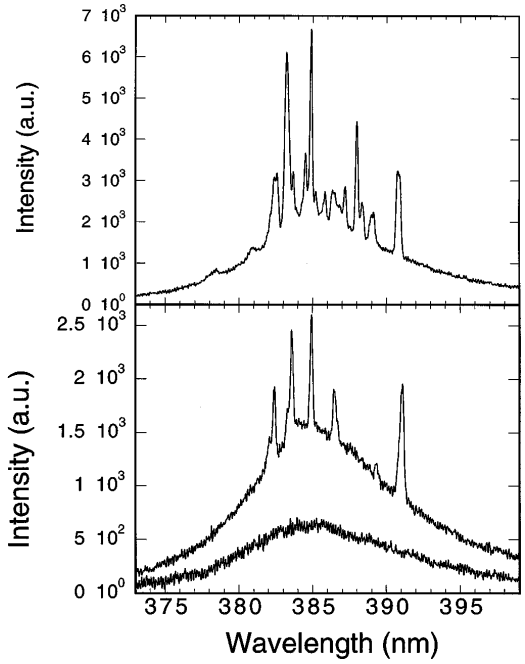

FIG. 4. Emission spectra when the excitation area is (from bottom to top) 980, 1350, and 1870 μm^2. The excitation intensity is 1012 kW/cm^2.

with the observation angle. Since different laser cavities formed by multiple scattering could have different output directions, lasing modes observed at different angles are different.

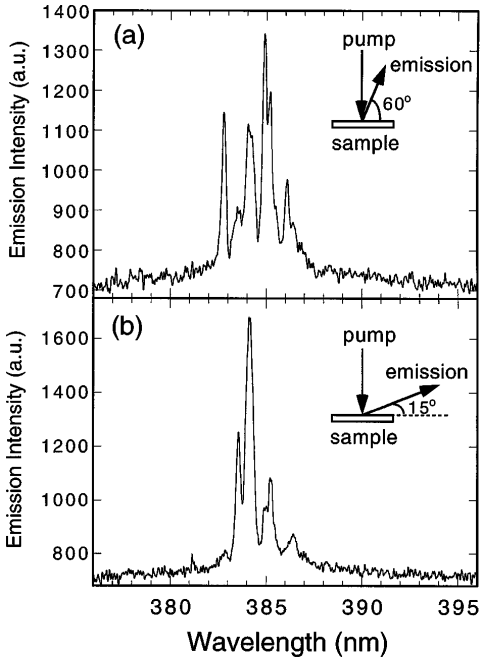

FIG. 5. Spectra of laser emission into two directions: (a) 60° from the sample surface, (b) 15° from the sample surface. The excitation intensity is 1188 kW/cm^2. The excitation area is about 1130 μm^2. The insets illustrate the experimental configuration.

The above experiments were repeated on GaN powder with an average particle size of 100 nm. Similar lasing phenomena were observed.

Finally, we would like to comment that our work is totally different from the previous work on powder laser [20,21]. Lasing was observed in Nd-doped laser crystal powder whose particle size was more than 10 μm. Since the particle size was much larger than the wavelength, a single particle could serve as a resonator. For the semiconductor powder we have studied, the particle size is less than the emission wavelength, according to the scanning electron microscope pictures. Thus a single particle is too small to serve as a laser resonator. Instead, laser resonators are formed by recurrent light scattering [22].

In summary, we have observed, for the first time, random laser action with coherent feedback in semiconductor powder. This observation provides direct evidence for the existence of recurrent scattering of light, which is the key ingredient in Anderson model of photon localization. We have demonstrated that recurrent light scattering can provide resonant feedback for lasers. Our study of random lasers may help the understanding of galaxy masers which are based on the same feedback mechanism [23,24].

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