Dynamics of random laser and coherent backscattering of light from ZnO amplifying random medium

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We investigated the dynamics of coherent backscattering cone and random laser in a disordered gain medium of ZnO powder. Theoretical fit to the coherent backscattering cone yields a transport mean free path l approximately equal to the probe wavelength λ , indicating that the scattering process is in the strong scattering regime. Supernarrow emissions from the laser are accompanied by enhancement and sharpening of the coherent backscattering cone. This enhanced and sharpened backscattering cone returns to a normal profile within 10–20 ps, indicating an ultrafast gain lifetime in the medium, which is in good agreement with the time-resolved measurement of the random laser emissions. © 2007 American Institute of Physics. [DOI: 10.1063/1.2824395]

Recent observations of random lasing (or spectral super-narrowing with a spectral width of a few angstroms) in polycrystalline ZnO and polymer films have generated considerable interests.^{1–7} In ZnO case, well-defined laser modes and spatial confinement of light have been observed and, therefore, the emissions are likely to be true laserlike emissions.^{1–4} In polymer films, photon statistics measurements confirm the coherence and stimulated nature of the sharp emission peaks, and Fourier transform of the emission spectra also verified the existence of random laser cavities.^{5–7} Observation of coherent backscattering cone (CBC) is the true test of random scattering. The angular width of the backscattering cone can then be used to determine whether the scattering process is in the strong or weak localization regimes.⁸ Although coherent backscattering has been measured in amplified random media,^{9–11} the systems studied were only in the weakly scattered regime.

Little experimental work has been done on random laser dynamics, and most of the investigations that have been conducted have been in solution.^{12,13} Recently, Cao and co-workers have observed delayed emissions, relaxation oscillation, and fast decay in random lasing in closely packed ZnO nanoparticles.^{14–16} However, no time-resolved measurements of the coherent backscattering cone under lasing conditions, which directly probe the optical gain and random scattering dynamics, have been reported. There have been a number of theoretical predictions and simulations of random laser dynamics. These models predict spectral narrowing, large reduction in emissions lifetime, delayed emissions, and relaxation oscillation, similar to behaviors of conventional lasers with optical cavities.^{17–19} In this letter, we report time-resolved measurements of the coherent backscattering cone due to random lasing in ZnO powder. We also observed enhancement and sharpening of the CBC. Our results showed that photons undergo stimulated amplification by random scattering to form optical cavities with coherent feedbacks.

We used pump-probe measurements to study the dynamics of the coherent backscattering peak. A pump beam at 267 nm was generated from sum frequency mixing between 800 and 400 nm femtosecond laser pulses from a Ti:sapphire regenerative amplifier (~200 fs pulse width). This 267 nm

pump beam was used to excite the ZnO powder sample to generate the stimulated emissions or laser. It is well known that in generating supernarrow spectral emissions (i.e., spectral width of less than 1 nm), careful control of the excitation spot size and intensity is essential. Therefore, micrometer-controlled knife edges and a variable neutral density filter in the pump beam path were used to control the spot size and light intensity. The probe beam was generated by focusing the 400 nm beam on a fused silica plate to produce a continuum and a narrow band filter to select the 387 nm probe. This 387 nm probe corresponded to the peak of the stimulated emissions when the ZnO was under moderate or high excitation. The 387 nm beam therefore probed the gain dynamics accompanying the random laser. The angular distribution of the 387 nm backscattered light was measured by a detector with a pin hole in front mounted on a scanning stage. A narrow band pass filter was used to detect only the coherent backscattered light of the probe beam at 387 nm. The profile and intensity of the backscattering peak for excitation above and below the “lasing” threshold could then be measured. We can also measure the stimulated emission spectrum simultaneously by collecting the light using an optical fiber that was coupled to a spectrometer with a charge coupled device. In this way, we could monitor precisely the excitation intensity at which spectral supernarrowing occurs and the concurrent change in the coherent backscattering peak profile. By varying the time delay between the pump and the probe beams, the time evolution of the coherent backscattering peak could be measured. Picosecond time-resolved photoluminescence (PL) of the individual random lasing modes was also measured using a streak camera coupled to a spectrometer to complement the time-resolved pump-probe measurements.

The ZnO samples were in pellet form with thicknesses of about 1 mm. These pellets were made by compressing the ZnO powder under high pressure. A transmission electron microscopy image of the ZnO powder indicated that there were ZnO crystalline grains with sizes from a few tens to a few hundreds of nanometers in diameter. This small grain size ensured that the sample was highly scattered and the transport mean free path l was small.

Figure 1 shows the PL spectra at a constant excitation intensity as a function of the excitation area (i.e., a micron in length and b micron in width), where $b=75\text{ }\mu\text{m}$ was kept

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FIG. 1. The stimulated emission spectra of ZnO powder as a function of the excitation area with a fixed peak pump intensity of $\sim 2.30 \text{ GW/cm}^2$.

constant and a was allowed to vary. The spectra clearly show that the emission spectrum changes from a broad and typical PL (bandwidth of $\sim 20 \text{ nm}$) with a very small excitation area to a PL with supernarrow emissions when pumped above a certain critical excitation area. These supernarrow emissions are indication of the presence of random lasers with coherent feedback.¹⁻⁴ The coherent feedback is due to random scattering from the crystalline grains in the highly disordered ZnO powder. Laserlike modes arising from strong scattering formed closed loop paths that act as laser ring cavities. Increasing the excitation area increases the number of emitted laser modes. With the excitation area fixed and the excitation intensity varied, similar emission spectra to those shown in the Fig. 1 were obtained. Under high excitation or large-area excitation, the lasing modes merged into a single simulated emission band, as shown in Fig. 1, with an excitation area of $97.5 \times 75 \mu\text{m}^2$. These are the typical lasing spectra observed in the previous random scattering experiments.¹⁻⁷

As mentioned previously, a true test to determine if a system is undergoing random scattering is the observation of the CBC.⁸ Figure 2(a) shows the backscattered light at 387 nm as a function of the angle, which clearly shows the CBC. This backscattering cone enables us to estimate the transport mean free path l using the three-dimensional coherent backscattering model.²⁰ The solid curve in Fig. 2(a) is the fit using this model yielding a transport mean free path l approximately equal to 0.8λ , which is close to the strong scattering regime. This value of l is substantially shorter than those observed in ZnO film² but consistent with reports of a ZnO powder deposited on a glass substrate by electrophoresis.¹

To understand the behavior of the coherent backscattering process and the excited state lifetime at the stimulated emission peak of $\sim 387 \text{ nm}$ when the ZnO random medium undergoes random lasing, we used the pump-probe technique to study the coherent backscattering cone. Figure 2(b) shows the backscattering cone at 387 nm at zero and 10 ps time delays relative to the 267 nm pump beam. The CBC is enhanced and sharpened at zero time delay. In this case, the full width at half maximum of the backscattering cone is reduced to 3° compared with $\sim 7.2^\circ$, as shown in Fig. 2(a), for the same system without the band gap excitation to produce the

FIG. 2. CBC at 387 nm. (a) Angular dependence of the backscattering peak for the probe beam only and fitted with the three-dimensional coherent backscattering equation to obtain a transport mean free path $l=0.8\lambda$. (b) Enhancement and sharpening of the CBC when pumped with a 267 nm beam above the random lasing threshold at zero time delay relative to the probe beam. The backscattering cone is considerably noisier in case (b) than in case (a) because of the additional fluctuations from the high intensity pump beam and the pump-induced PL.

gain. From this reduced width, we can estimate the depth of the gain region (z_l) using the relation $z_l = \lambda / 2\Delta\theta \approx 3.7 \mu\text{m}$.²¹ This value of z_l is about ten times that of l , which is within the range of theoretical predictions²¹ and seems to be reasonable considering the strong optical pumping in our case. At 10 ps time delay, the backscattering cone width returns to a value very similar to the unpumped case [i.e., Fig. 2(a)], indicating an ultrafast gain lifetime. Since the amount of amplification experienced by the probe beam is dependent on the length of the gain medium on which it travels, it will depend on the scattering of the probe photons with the ZnO grains in the sample. Since light from the center of the backscattering cone experiences the largest scattering (i.e., the longest path length for amplification), the CBC is thus enhanced and sharpened when the pump beam is sufficiently strong to produce gain in the active medium. This phenomenon of an enhanced backscattering cone was also observed in other active media, such as Ti:sapphire powder and a dye-particle mixture under a weak scattering regime, and its behavior above and below the laser threshold is also similar.^{9,10} In particular, we did not observe a runaway growth of the backscattering cone above the laser's threshold, probably due to ultrafast population decay above the laser threshold.¹¹ We must also emphasize that the sharp peak seen at the center of the backscattering cone at zero delay is not an artifact such as a speckle effect. The CBC measured from the average of a number of scans and the picosecond decay of the CBC (see Fig. 3) clearly shows that the enhanced and sharpened CBC is due to optical gains introduced by external pumping.

Figure 3 shows the decay of the $\sim 387 \text{ nm}$ scattered probe beam intensity at the center of the backscattering cone as a function of the time delay. The figure's inset shows the

FIG. 3. Picosecond decay of the CBC from the 387 nm probe beam at the backscattered angle $\theta=0^\circ$. The inset shows the corresponding time-integrated emission spectrum, which clearly indicates single mode random laser emissions at ~ 387 nm, and the probe continuum spectrum nicely overlaps with the single mode emission

corresponding PL and the probe beam spectra. A single sharp laser mode superimposed on the broad PL background is clearly seen, and the probe beam is exactly at the same wavelength as the lasing mode. Thus, the 387 nm beam probes the excited state gain lifetime. An ultrafast decay time of ~ 20 ps is clearly seen in Fig. 3. This result is also consistent with the time-resolved PL shown in Fig. 4 where emission spectra pumped to just above the threshold exhibit random lasing. A single mode at different time delays is shown. This time-resolved PL shows that laserlike, supernarrow emissions at ~ 385 nm disappeared completely within 30 ps, leaving only the broad spontaneous emissions at longer time delays. The figure's inset shows more clearly the detailed PL decay dynamics at the 385 nm lasing peak and the 377 nm spontaneous emission wavelength. The spontaneous emissions at 377 nm show a lifetime of ~ 100 ps, which is typical of PL lifetimes for ZnO films and nanostructures.²² On the other hand, the single mode random lasing emission at 385 nm has a decay lifetime of less than 30 ps, which is close to the time resolution limit of our streak camera system. The fact that both the pump-probe and time-resolved PL measurements of the supernarrow emissions at ~ 385 nm have ultrafast lifetimes clearly demonstrates that this sharp emission peak is indeed due to stimulated emissions (i.e., lasing).

FIG. 4. Normalized time-resolved single mode random laser spectra showing the relaxation dynamics of the laser emissions and the PL background. The inset shows the decay of the spontaneous emission at 377 nm and the single mode laser peak at ~ 385 nm. The dotted line shows only the decay dynamics of the stimulated laser emission obtained by subtracting the slow spontaneous PL background from decay curve (a), which resulted in a single exponential decay with a decay time constant of ~ 29 ps.

In conclusion, we investigated the random laser phenomenon in a disordered gain medium of ZnO powder. Supernarrow spectral lines (i.e., from lasing) were observed under controlled excitation intensity and/or excitation spot size. The CBC due to the random multiple scatterings by the ZnO crystalline grains yields a transport mean free path l approximately equal to λ , which is close to the strong scattering regime. Laser emissions together with the dramatic enhancement and sharpening of the CBC are concurrently observed. Since light from the center of the backscattering cone experiences the largest scattering (i.e., the longest gain length), this sharpening and enhancement of the coherent backscattering peak are thus consistent with the random laser model, which postulate that the supernarrow peak is due to amplification and stimulated emission of photons in the random gain medium. The time-resolved PL and the coherent backscattering peak show that the gain lifetime above the lasing threshold is less than 30 ps, which is consistent with the suggestion that the supernarrow peaks are due to stimulated emissions. Our results showed that photons undergo stimulated amplification in the gain medium by random scattering to form optical cavities with coherent feedback and to produce supernarrow emissions, i.e., random lasers.

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