Far-field characteristics of random lasers

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(Received 19 November 1998)

We report on experimental observation of the far-field intensity and mode distributions of random lasers. Laser emission from highly disordered semiconductor polycrystalline thin films could be observed in all directions. The angle dependence of the laser output from the edge of the film is different from that of the laser emission scattered out of the surface of the film. More lasing modes are observed from the surface of the film than from the edge of the film. A qualitative explanation of the experimental results are presented based on the laser cavities formed by optical scattering being located in the plane of the films. [S0163-1829(99)04623-8]

I. INTRODUCTION

Weak scattering of light is detrimental to laser action since it removes photons from the lasing mode of a conventional cavity. However, when optical scattering is sufficient, it may facilitate lasing by forming resonators.[1–5] Specifically, when the scattering mean-free path approaches the optical wavelength, the light may return to a scatter from which it was scattered before, and thereby forming closed loop paths. If the amplification along the closed loop paths is strong enough, these closed loops could serve as ring cavities for light, and the system might lase in the modes of these localization cavities. This kind of laser is called a ''random laser.''

Recently, we have observed such random lasers. The laser action occurred in highly-disordered ZnO and GaN powder and polycrystalline thin films.[6] Scanning electron microscopy images indicate that the ZnO films consist of many irregularly shaped grains with sharp interfaces. At low-pump power, the emission spectrum consists of a broad spontaneous emission peak. When the excitation intensity exceeds a threshold, very narrow peaks emerge in the emission spectrum, and the total emission intensity increases much more rapidly with the pump power.

For traditional lasers with well-defined cavities, the far-field intensity and mode distributions are determined by the cavity configuration and gain distribution. It is interesting to study the far-field intensity and mode distributions of random lasers, since it is expected to be quite different from that of traditional lasers. In this paper, we report our experimental observation of the far-field intensity and mode distributions of the random lasers.

II. EXPERIMENT

Thin films of (0001) ZnO were deposited on (0001) sapphire substrates by pulsed laser ablation. A detailed description of the growth procedure and the structural characterization of the films have been given elsewhere.[7,8] The thickness of the films is about 300 nm. A frequency-tripled mode-locked Nd:YAG laser (355 nm, 10 Hz repetition rate, 15 ps pulse width) was used to optically pump the samples. The emission from the samples was collected by a fiber bundle and directed to a 0.5-meter spectrometer with a cooled charge coupled device array. The tip of the fiber bundle can be moved along a circular trail centered at the excitation spot on the film. The distance from the fiber tip to the excitation spot is about 1 cm.

First, we measured the far-field intensity and mode distributions in the plane of the film. The experimental configuration was shown in the inset of Fig. 1. The pump beam was focused by a cylindrical lens to a stripe with a width of ~ 40 μm on the ZnO film at normal incidence. The stripe length could be varied by an adjustable slit. One end of the

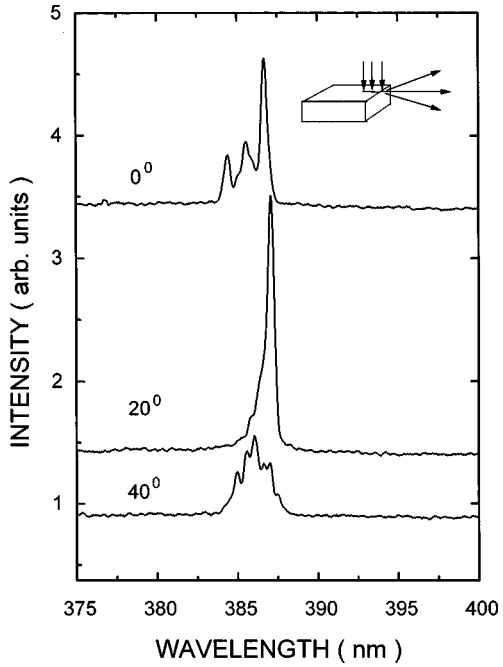

FIG. 1. Side emission spectra observed from different angles (0°, 20°, and 40°) in the plane of the film. The excitation intensity is 383 kW/cm^2. The excitation stripe length is 155 μm. The inset shows the experimental configuration.

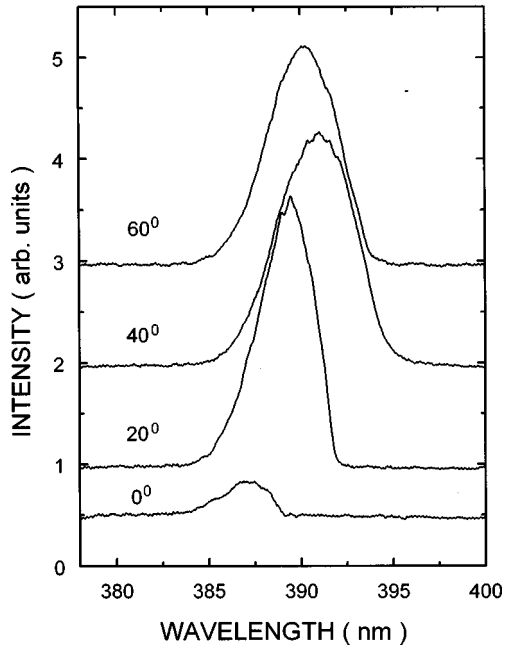

FIG. 2. Side emission spectra observed from different angles (0°, 20°, and 40°) in the plane of the film. The excitation intensity is 381 kW/cm^2, and the excitation stripe length is 1 mm.

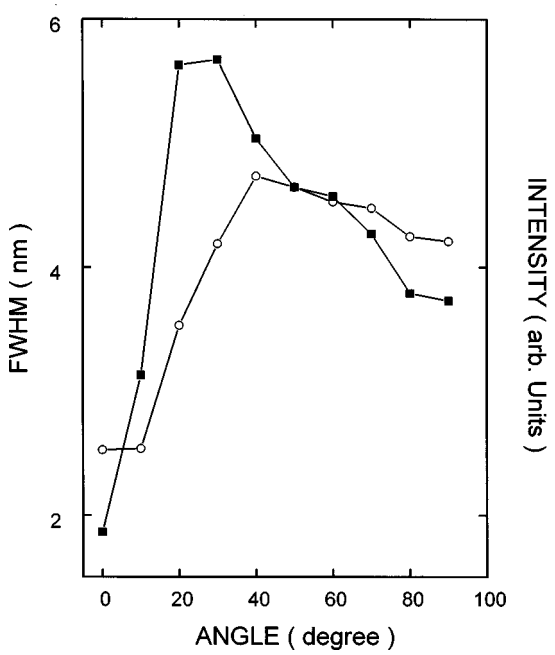

FIG. 3. The intensity (squares) and FWHM (circles) of the laser spectra as a function of the observation angle in the plane of the film. The excitation intensity is 381 kW/cm^2, and the excitation stripe length is 1 mm.

stripe was close to the cleaved edge of the sample. The laser emission from the edge of the film was measured.

Figure 1 shows the side emission spectra at various observation angles in the plane of the film. The excitation intensity is 383 kW/cm^2. At 0° (normal to the cleaved edge of the sample), the emission spectrum consists of three discrete peaks. However, at 20°, the spectrum has only one main peak. At 40°, six peaks emerge in the spectrum. Thus, the laser emission spectrum varies drastically with the observation angle.

As we increased the stripe length, more lasing peaks emerged in the emission spectrum. Eventually, there were so many lasing peaks that they could no longer be well resolved. Instead, they merged into a single broad peak. Figure 2 shows the laser emission spectra at various observation angles, when the excitation stripe is 1 mm long. At different observation angles, the center wavelength and the full width at half maximum (FWHM) of the broad peak are different. The shift of the peak wavelength is ~ 4.5 nm when the observation angle changes from 0° to 40°. Figure 3 shows the emission intensity and linewidth as a function of the observation angle. The maximum emission intensity is at 30°, while the widest linewidth is at 40°. We have repeated the same measurement with several samples. We found the maximum emission intensity and linewidth were different for different samples, and they also occurred at different angles. In another word, the far-field intensity and mode distributions in the plane of the film are rather random.

Next we measured the laser emission spectra from the sample surface at different observation angles. The excitation beam was focused by a spherical lens to a circle of ~ 50 μm in diameter on the sample surface at normal incidence. As shown in Fig. 4, the laser modes are basically the same and their intensities vary slightly when the observation angles are 0°, 20°, and 40°. Note, 0° corresponds to the direction normal to the substrate plane. As we increased the pump power, more and more lasing peaks emerged in the emission spectra, and eventually merged into a single broad peak. Figure 5 shows the laser emission spectra when the excitation intensity is 524 kW/cm^2. We can see that the center frequency, the linewidth, and the intensity of the broad lasing peak are

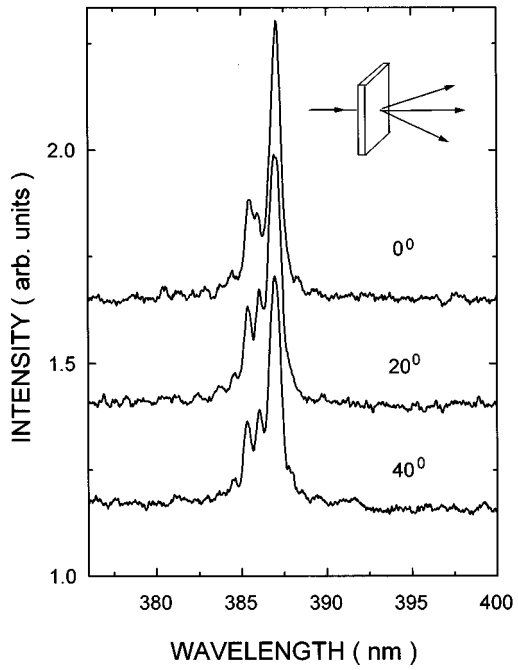

FIG. 4. Surface emission spectra observed at different observation angles (0°, 20°, and 40°) in the plane normal to the film. The excitation intensity is 382 kW/cm^2.

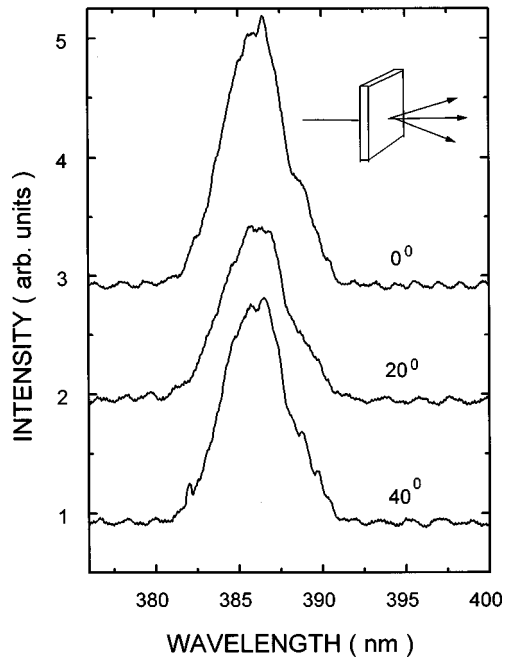

FIG. 5. Surface emission spectra observed at different observation angles (0°, 20°, and 40°) in the plane normal to the film. The excitation intensity is 504 kW/cm^2.

almost independent of the observation angles.

Finally, we studied the angle dependence of the laser emission in the plane perpendicular to both the cleaved edge of the sample and the substrate plane. The excitation area is a stripe close to the edge of the film. As shown in Fig. 6, at 0° (normal to the cleaved edge of the sample), the emission spectrum has two main lasing peaks. At 20°, more peaks

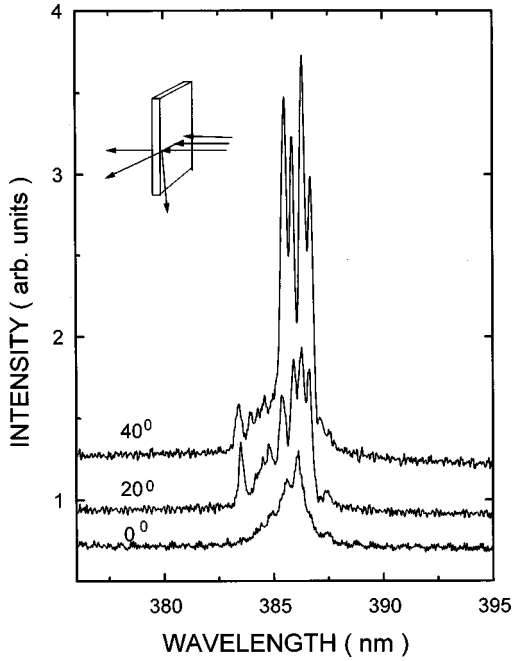

FIG. 6. Laser emission spectra observed at different angles in the plane perpendicular both to the cleaved edge of the sample and the substrate plane. The excitation intensity is 382 kW/cm^2. The inset shows the experimental configuration.

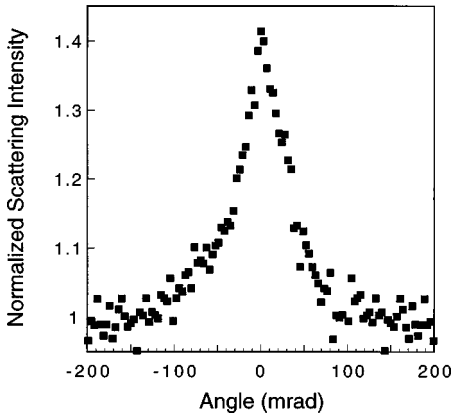

FIG. 7. Measured backscattering intensity as a function of angle. The ZnO film is about 9-μm thick.

emerge in the emission spectrum. At 40°, the number of lasing peaks does not increase any more. However the intensities of those peaks increase.

III. DISCUSSION

The highly disordered structure of ZnO polycrystalline films results in strong optical scattering. We characterized the scattering mean-free path l using coherent backscattering (CBS).[9–11] The ZnO film used in the CBS experiment was $\sim$9 μm thick. To avoid absorption, the frequency-doubled output ($\lambda = 410$ nm) of a mode-locked Ti:Sapphire laser (76 MHz repetition rate, 200 femtosecond pulse width) was used as the probe light. Figure 7 shows the measured backscattering cone of the ZnO film. From the angle of cusp, we estimated that the scattering mean-free path is about 2.6λ, after taking into account the finite thickness of the ZnO film.[12]

Because the scattering mean-free path is on the order of ZnO emission wavelength, closed loop paths for light could be formed through multiple scattering in the ZnO films. There are many such loop paths in the films. However, along different loop paths, the probability of a photon scattered back to its original point is different. In other words, the cavities formed by optical scattering have different loss. On the other hand, the ZnO films have rather large optical gain, e.g., the gain coefficient is over 20 cm^{-1} at a fluence of 5 μJ/cm^2. Under optical pumping, as the pump power increases, the gain reaches the loss first in the low-loss cavities. Thus, laser oscillation occurs in these cavities, and the lasing frequencies are determined by the cavity resonances. The laser emission from these resonators results in discrete narrow peaks in the emission spectrum. As the pump power increases further, the gain increases and it reaches the loss in the lossier cavities. Laser oscillation in those resonators add more discrete peaks to the emission spectrum. Eventually at very high pump power there are so many lasing peaks that they could no longer be distinguished from each other. Instead they merge into a single broad peak in the spectrum.

For ZnO thin films, since the optical scattering mean-free path is close to the film thickness but is much smaller than the lateral size of the excitation area, the laser cavities formed by optical scattering are located in the plane of the film. At a fixed pump intensity, with an increase of the excitation area, more lasing peaks emerge in the emission spec-

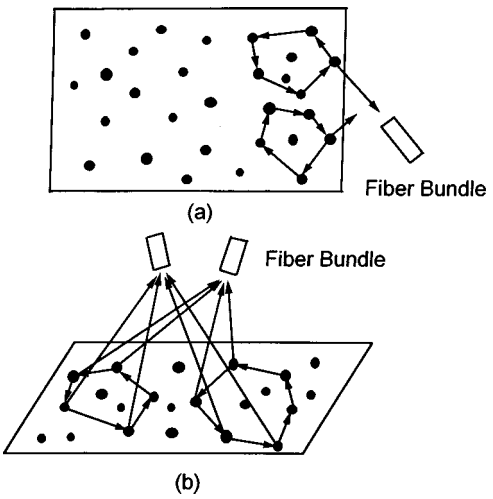

FIG. 8. A schematic diagram showing the random laser cavities in the ZnO thin film. (a) Laser output from the edge of the sample; (b) laser emission scattered out of the surface of the film.

tra, because there are more closed loop paths for light in a larger excitation area. Eventually, these discrete lasing peaks merge into a single broad peak. On the other hand, when the excitation area is smaller than a critical size, laser action does not occur, because the loop paths are too short and the amplification along the closed loops is not high enough to achieve lasing. Hence, the lasing threshold intensity increases as the excitation area decreases.

Figure 8 is a schematic diagram showing the random laser cavities in the ZnO film, and the laser emission from both the edge and the surface of the sample. The laser output from different cavities may go to different directions in the plane of the film. Hence, only the laser output whose direction is the same as the observation direction can be collected by the fiber bundle. That is why the laser emission spectra change drastically at different observation angles, as shown in Figs. 1 and 2.

Since the film thickness is close to the optical scattering mean-free path, the laser emission can easily be scattered out of the surface of the film by intracavity scatters. The scattering direction of the laser light may be different for different scatters. Therefore, the scattered laser light from each cavity could be collected by the fiber bundle at all observation angles. That is why in Figs. 4 and 5 the laser modes observed from the sample surface are almost the same at different observation angles.

With the experimental configuration shown in the inset of Fig. 6, when the observation angle increases, we start collecting laser emission scattered out of the surface of the film. That is why more laser modes emerge in spectra at larger observation angles.

For comparison, we have also measured the emission spectra from the edge of the sample below the lasing threshold. As shown in Fig. 9, the emission spectrum has a single broad peak originated from spontaneous emission. Contrary to the behavior of laser emission, the spontaneous emission peak does not shift in wavelength as we change the observation angle in the plane of the film. This is expected because the spontaneous emission frequency is determined by the band-gap energy regardless of the light path in the film. This observation confirmed that the data shown in Figs. 1–6 correspond to random laser emission, whose far-field mode distribution depends on the closed loop paths in the film.

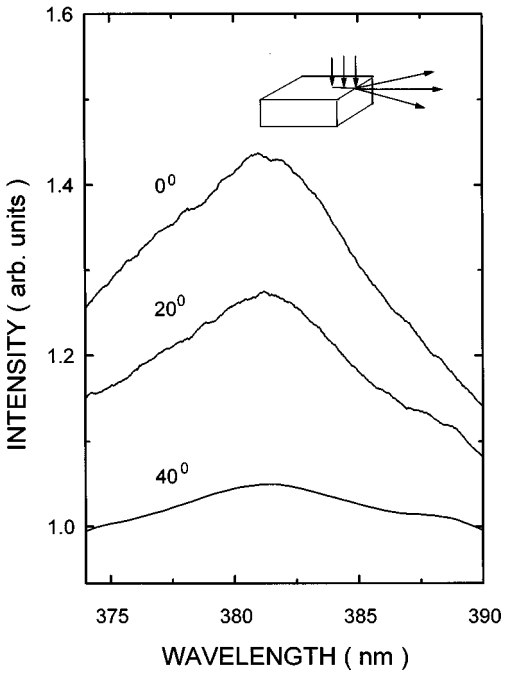

FIG. 9. Spontaneous emission spectra from the edge of the sample, observed at different angles (0°, 20°, and 40°) in the plane of the film.

Finally, we would like to comment that if the film thickness is much larger than the scattering mean-free path, the laser cavities formed by optical scattering are no longer confined in a plane. In this case, the far-field intensity and mode distributions from the film surface should be the same as those from the edges of the film.

IV. CONCLUSION

In summary, we have observed the far-field intensity and mode distributions of random lasers in highly disordered ZnO polycrystalline thin films. For the laser output from the edge of the film, both intensity and modes strongly depend on the observation angle in the plane of the film. However, for the laser emission scattered out of the surface of the film, the modes are nearly angle independent, while the intensity varies slightly with the observation angle. We have presented a qualitative explanation for the observed far-field characteristics of random lasers. Since the optical scattering mean-free path is close to the film thickness but is much smaller than the lateral size of the excitation area, the laser cavities formed by optical scattering are located in the plane of the film. The laser output from different cavities may go to different directions in the plane of the film. On the other hand, the laser light could be scattered out of the surface of the film by intracavity scatters into all directions.

ACKNOWLEDGMENT

This work was partially supported by the National Science Foundation under Grant No. ECS-9877113.

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