Effect of external feedback on lasing in random media

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We have studied the effect of external feedback on random laser action in ZnO polycrystalline thin films. Reinjection of light into scattering-formed cavities strongly influences modes, intensity, and threshold of random lasers. We have compared the effect of external feedback from the side of the film and that from the film surface. Our study opens the possibility of controlling random laser frequencies by external feedback.
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Interfaces have strong influence on diffusion transport of light through highly disordered media. As a result of discontinuity in refractive index at sample interfaces, light that attempts to exit through sample interfaces could be reflected. The photons reinjected into the medium execute a second, independent random walk before once again attempting to exit. Thus the average time a photon spends in the medium, the average path length it travels, and the average number of scattering events it undergoes before exiting are all drastically increased due to interface reflections. The significant contribution of interfaces is first noticed in the optical memory effect.[1,2] Subsequent studies have illustrated the effect of light reinjection on coherent backscattering, pulse transmission, frequency-, and time-dependent speckle correlations.[3–6]

The above studies are on ''passive'' random media. In an ''active'' random medium, interface reflection and external feedback play important roles in light amplification and lasing. It is predicted that internal reflection by a large index mismatch at sample boundaries may facilitate lasing.[7] Laser-like emission has been observed from laser dye solution containing TiO_2 microparticles.[8,9] Interface reflection and external feedback reinject photons into the gain regime, leading to a reduction of the threshold for linewidth collapse.[10] However, the spectral linewidth of the laser-like emission is several nanometers, and no individual lasing modes are observed.

Recently we have observed lasing with resonant feedback in highly disordered ZnO powder and polycrystalline films.[11] Above the threshold, discrete lasing modes whose spectral linewidth is less than 0.3 nm emerge in the emission spectra. Since the scattering mean free path approaches the emission wavelength, closed loop paths for light could be formed through multiple scattering. Under optical pumping, the ZnO samples exhibit quite large optical gain. When the optical amplification along a scattering-formed loop exceeds the loss, laser oscillation occurs in the loop, and lasing frequencies are determined by the cavity resonances. In this letter, we study the effect of external feedback on the modes of random lasers.

ZnO films (300–350 nm thick) are deposited on amorphous fused silica or sapphire substrates by pulsed laser ablation. A pulsed KrF excimer laser (248 nm) was used to ablate a hot pressed ZnO target in an ultrahigh vacuum chamber with a base pressure of 1×10^{-8} Torr. Films were deposited in an oxygen partial pressure of 10^{-5}–10^{-4} Torr at substrate temperature of 500–700 °C. A detailed description of the growth procedure and the structural characterization of the films have been given elsewhere.[12,13] The high resolution transmission electron microscope (HRTEM) images indicate that the films consist of many irregularly shaped grains whose sizes vary from 20 to 150 nm. In the photoluminescence experiment, the films are optically excited by the frequency-tripled output of a mode locked Nd:YAG laser (355 nm, 10 Hz repetition rate, 15 ps pulse width). The pump beam is focused to a stripe (40 μm wide) with normal incidence. The length of the stripe can be varied continuously from a few microns to hundreds of microns by an adjustable slit. One end of the stripe is close to an edge of the sample. Emission from the edge of the film is directed to a spectrometer with a cooled CCD array.

To study the effect of external feedback, an aluminum mirror is placed near the sample side opposite to the excitation stripe. The mirror plane is perpendicular to the film and the excitation stripe, as shown in the inset of Fig. 1. The lateral size of the sample is $\sim$3 mm. Figure 1 shows the laser emission spectra when the distance from the mirror to the film edge opposite to the excitation stripe is 0, 80, and 140 μm. For comparison, laser emission spectrum without mirror is also plotted. As the mirror is moved closer to the film edge, laser intensity is increased. Some lasing modes emerge, and some modes disappear.

Figure 2 shows the incident pump power required to reach lasing threshold as a function of distance from the mirror to the film edge opposite to the excitation stripe. The lasing threshold pump power is reduced to one half in the

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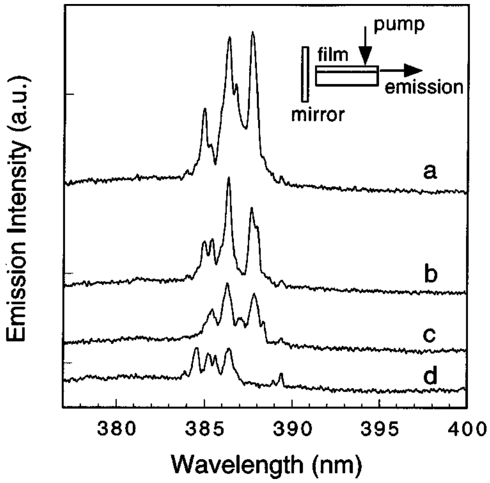

FIG. 1. Laser emission spectra when the distance from the mirror to the film edge opposite to the excitation stripe is (a) 0, (b) 80 μm, and (c) 140 μm, (d) is the laser emission spectrum without mirror. The excitation stripe is 100 μm long. The incident pump intensity is 14.5 $\mu J/cm^2$. The inset illustrates the experimental configuration.

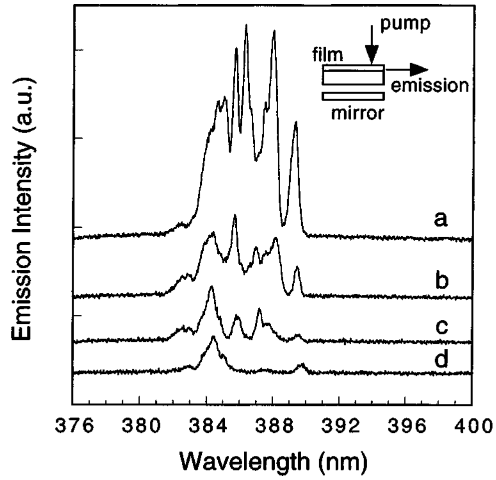

FIG. 3. Laser emission spectra when the distance from the mirror to the substrate surface is (a) 0, (b) 120 μm, and (c) 240 μm, (d) is the laser emission spectrum without mirror. The excitation stripe is 100 μm long. The incident pump intensity is 14.5 $\mu J/cm^2$. The inset illustrates the experimental configuration.

presence of feedback. When the distance from the mirror to the film edge is more than 800 μm, the lasing threshold pump power with the mirror is almost the same as that without the mirror.

Next, we put the mirror underneath the sample. The experimental configuration is shown in the inset of Fig. 3. The mirror plane is parallel to the film. The laser emission spectra changes drastically with the distance from the mirror to the substrate surface (see Fig. 3). When the mirror is moved closer to the substrate, more lasing modes emerge in the spectra, and laser intensity increases.

The effect of external feedback on spectral characteristics of traditional semiconductor lasers has been studied extensively.[14,15] The whole system can be modeled as two cavities formed by three mirrors. The external cavity can be treated as a wavelength-dependent end mirror of the primary cavity. However, there are no traditional cavities in our samples. Thus the three mirror model cannot be used to explain our experimental results.

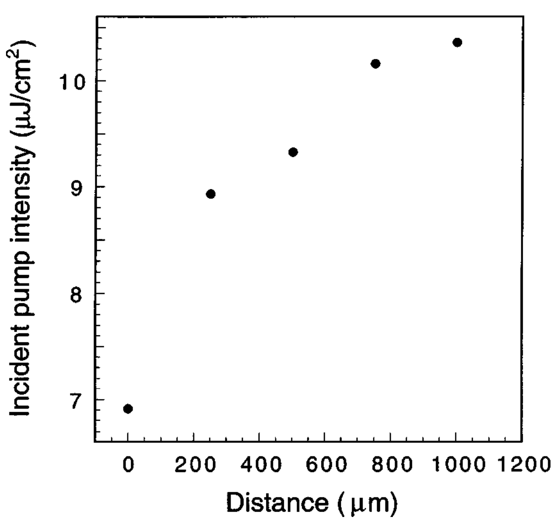

FIG. 2. Incident pump power required to reach lasing threshold as a function of distance from the mirror to the film edge opposite to the excitation stripe. The length of the excitation stripe is 100 μm.

In ZnO polycrystalline films, there are many closed loop paths for light formed by optical scattering. In those loops where loss is less than optical amplification, laser oscillation occurs. The lasing modes in the emission spectra are from different loops formed by scattering. Since the film thickness is on the order of the scattering mean free path but much smaller than the lateral size of the excitation area, the laser cavities formed by scattering are in the plane of the film.[16] Due to the highly disordered structure of ZnO films, emitted light and pump light are strongly scattered as they travel in the films. Eventually the emission exits the films through film surfaces or edges. When a mirror is placed near an edge of the film, the light which attempts to escape from the film edge is reflected by the mirror and reinjected into the film. Reinjection of ZnO emission into scattering-formed cavities reduces the ‘‘effective’’ cavity loss, while reinjection of pump light into the excitation area increases pump intensity. Thus the pump power required to reach lasing threshold decreases. In some cavities whose loss is too large to achieve lasing without external feedback, laser action may occur due to light reinjection. Laser emission from these cavities adds lasing peaks in the emission spectra. However, some cavities where lasing occurs with the help of external feedback might have spatial overlap with the cavities where lasing already occurred without feedback. Gain competition could suppress lasing in the latter cavities, and lead to the disappearance of some lasing modes in the presence of feedback.

When the mirror is placed underneath the sample, the pump light which transmitted through the film is reflected by the mirror and reinjected into the film. Hence, the pump intensity is increased. The increase of optical gain not only increased laser intensity, but also led to lasing in some scattering-formed cavities which could not lase before. Thus new lasing peaks emerged in the spectra. In addition, since the incident pump beam is focused onto the film, the reflected pump beam is defocused. The reflected pump beam spot on the film is larger than the incident pump beam spot. Thus the excitation area is also increased, leading to lasing in some scattering-formed cavities which are located outside

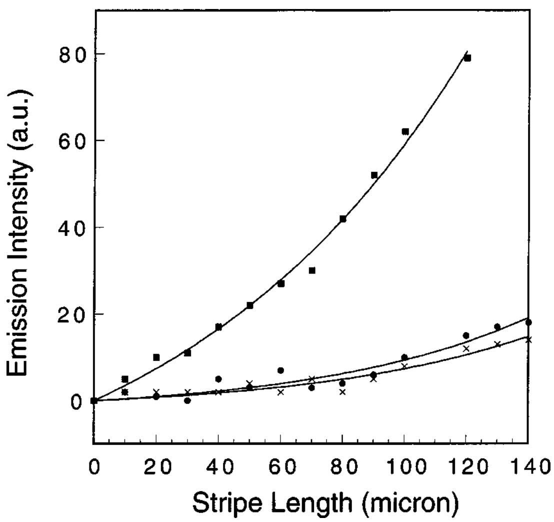

FIG. 4. Spectrally integrated intensity of ASE from the film edge as a function of the stripe length in three cases: without a mirror (circle), with a mirror at the sample side opposite to the excitation stripe (square), with a mirror underneath the substrate (cross).

(or partially outside) the incident pump beam spot but inside the reflected pump beam spot. Additional lasing peaks emerged in the emission spectra.

To compare the effect of external feedback in the above two configurations, we have measured the dependence of amplified spontaneous emission (ASE) intensity on the excitation stripe length below the lasing threshold. Figure 4 shows the spectrally integrated intensity of ASE from the film edge as a function of the stripe length in three cases: without a mirror, with a mirror at the film side opposite to the excitation stripe (see inset of Fig. 1), with a mirror underneath the substrate (see inset of Fig. 3). The incident pump intensity is chosen such that lasing occurs when the stripe is 160 μm long in all three cases. The dependence of ASE intensity on the stripe length in the presence of feedback from the substrate surface is nearly the same as that without feedback. In contrast, ASE intensity increases much more rapidly with the stripe length with feedback from the film edge than that with feedback from sample surface. This is because ASE which attempted to exit from the film edge is bounced back by the mirror, and part of it is reinjected into the gain regime. The reinjected ASE is amplified more as it traveled in the gain regime before another attempt of escape. An increase of the path length of photons in the gain regime results in an increase of ASE intensity. As the stripe became longer, more reflected ASE could be reinjected into the gain regime, and thus ASE intensity increased very rapidly with the stripe length. On the other hand, for ASE scattered out of the film–substrate interface, it could be reflected by the mirror underneath the substrate, and part of it is reinjected into the gain regime. However, since the film is rather thin, the path length of the reflected photons in the gain regime is quite short. Hence the reinjected ASE is not amplified more before escaping from the film surface. That is why a mirror underneath the substrate barely changes the dependence of ASE intensity on the stripe length.

This result suggests that the contributions of external feedback in the above two cases are different. A mirror placed underneath the substrate leads to an increase of pump intensity and excitation area. On the other hand, a mirror placed at the film side not only increases the excitation intensity and area due to reflection of pump light, but more importantly reinjects ZnO emission into the gain regime. The reinjected emission helps to achieve lasing in some scattering-formed cavities.

Similar effects of external feedback have been observed with thin films of ZnO powder with random crystalline orientation.

In summary, we have demonstrated that external feedback strongly influences modes, intensity, and threshold of random lasers. Since the ZnO polycrystalline films are thin, the effect of external feedback from the film side is different from that that of feedback from the film surface. Our study opens the possibility of controlling random laser frequencies by external feedback.

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