Ultraviolet lasing in resonators formed by scattering in semiconductor polycrystalline films

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A semiconductor laser whose cavities are ''self-formed'' due to strong optical scattering in highly disordered gain media is demonstrated. The lasers are made of zinc oxide polycrystalline films grown on amorphous fused silica substrates. Lasing occurs at an ultraviolet wavelength of ~380 nm under optical pumping. Actual images of the microscopic laser cavities formed by multiple scattering have been captured. These results suggest the possibility of using disordered semiconductor microstructures as alternative sources of coherent light emission. © *1998 American Institute of Physics.* [S0003-6951(98)00750-5]

Commercially available compound semiconductor lasers are made of single-crystalline films fabricated by heteroepitaxial growth on lattice-matched substrates. However, epitaxial growth techniques are not only expensive but are also very restrictive to the type of substrates which can be used. The substrate restriction and fabrication cost have limited the applications of semiconductor lasers. We have focused our research effort on the fabrication of semiconductor lasers by nonepitaxial growth of semiconductor materials on various substrates. In this letter, we report the demonstration of ultraviolet (UV) laser action in zinc oxide (ZnO) polycrystalline films grown on amorphous fused silica substrates by laser ablation.

These lasers are not traditional semiconductor lasers which have well-defined cavities. Instead, laser cavities are ''self-formed'' due to strong optical scattering in the polycrystalline films. Optical scattering has traditionally been considered detrimental to laser action since it induces loss. However, we will show that when the optical scattering is strong enough, it actually helps lasing by forming closed-loop paths for light and introducing feedback.[1,2]

ZnO films (300–350 nm thick) were deposited on amorphous fused silica substrates by 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 temperatures of 500–700 °C. A detailed description of the growth apparatus and growth procedure will be provided elsewhere.[3]

X-ray diffraction measurement indicates that the ZnO films are textured with the *c* axis oriented perpendicularly to the substrate plane. The full width at half maximum (FWHM) of the (0002) ω-rocking curve shows a mosaic spread of 0.9°–1°. In-plane x-ray Φ scans of the films exhibit random in-plane crystal alignment with respect to the substrates. This result is consistent with the fact that an amorphous substrate does not provide any preferred lateral growth direction on its surface.

The optical absorption measurement of the ZnO films indicates that the band edge of ZnO is ~0.05 eV lower than that epitaxially grown on a sapphire substrate.[4] In the photoluminescence experiment performed at room temperature, UV emission due to the transition from the conduction band to the valence band was observed. However, no green emission originated from deep levels could be detected after we successfully passivated the defect states in the films during the growth.[3]

In the absence of any fabricated mirrors, we observed lasing in the ZnO films. The samples were optically pumped by a frequency-tripled mode-locked Nd:YAG laser (355 nm, 10 Hz repetition rate, 15 ps pulse width). The pump beam was focused to a stripe or a spot on the film surface with normal incidence. Figure 1 shows the evolution of the side emission spectra as we increased the pump power. At low excitation intensity, the spectrum consists of a single broad spontaneous emission peak. As the pump power increases, the emission peak becomes narrower due to the preferential amplification at frequencies close to the maximum of the gain spectrum. When the excitation intensity exceeds a threshold, very narrow peaks emerge in the emission spectra. As the pump power increases further, more sharp peaks appear. The linewidth of these peaks is less than 0.4 nm, which is more than 20 times smaller than the linewidth of the amplified spontaneous emission peak below the threshold. Above the threshold, the integrated emission intensity increases much more rapidly with the pump power, as shown in the inset of Fig. 1(b). The emission above the threshold were measured to be strongly polarized [see the inset of Fig. 1(c)]. These data indicate that laser action has occurred in the ZnO films.

The characteristics of lasing in semiconductor polycrystalline films exhibits remarkable differences from that of a conventional laser. First of all, the laser emission from the ZnO films could be observed in all directions. The laser emission spectrum varied with the observation angle. Second, the pump intensity required to reach the lasing threshold depends on the excitation area. As the excitation area decreases, the lasing threshold density increases. Figure 2 shows the evolution of the emission spectra when the excitation area was varied at a fixed pump intensity. When the excitation area was increased, more lasing peaks emerged in the emission spectra. On the other hand, when the excitation area was reduced to below a critical size, laser oscillation stopped.

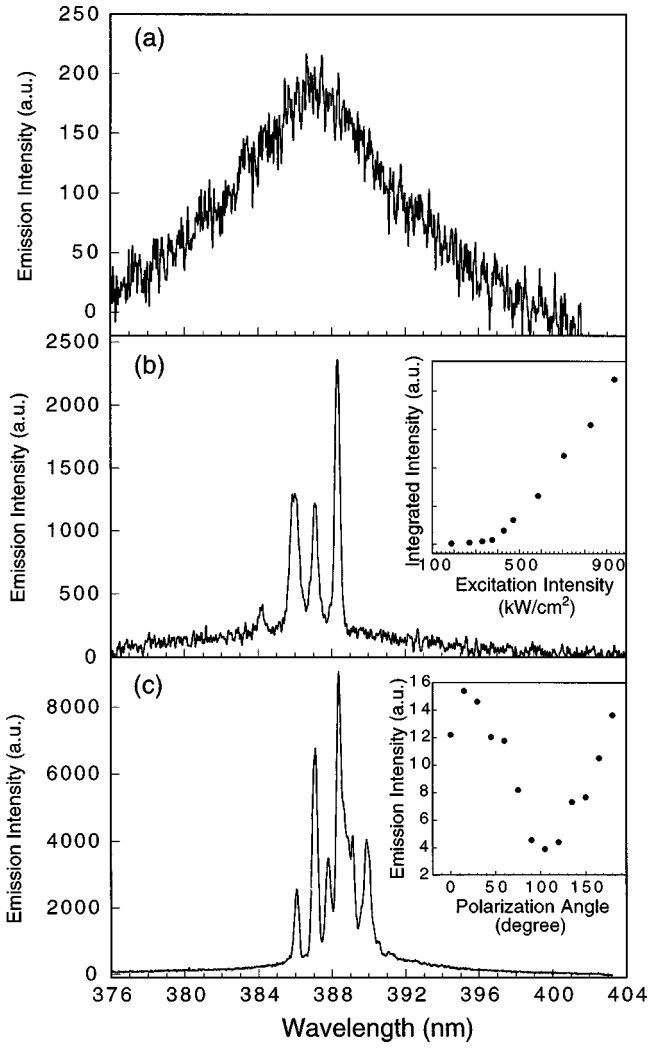

FIG. 1. Emission spectra when the excitation intensities are (a) 330 kW/cm^2, (b) 380 kW/cm^2, and (c) 600 kW/cm^2. The excitation area is a stripe of 100×40 μm. The emission is collected from the edge of the film. The inset of (b) shows the integrated emission intensity as a function of excitation intensity. The inset of (c) shows the intensity of the laser emission from the edge of the ZnO film as a function of polarization angle when the excitation intensity is 400 kW/cm^2. The intensity maximum corresponds to the polarization direction parallel with the film.

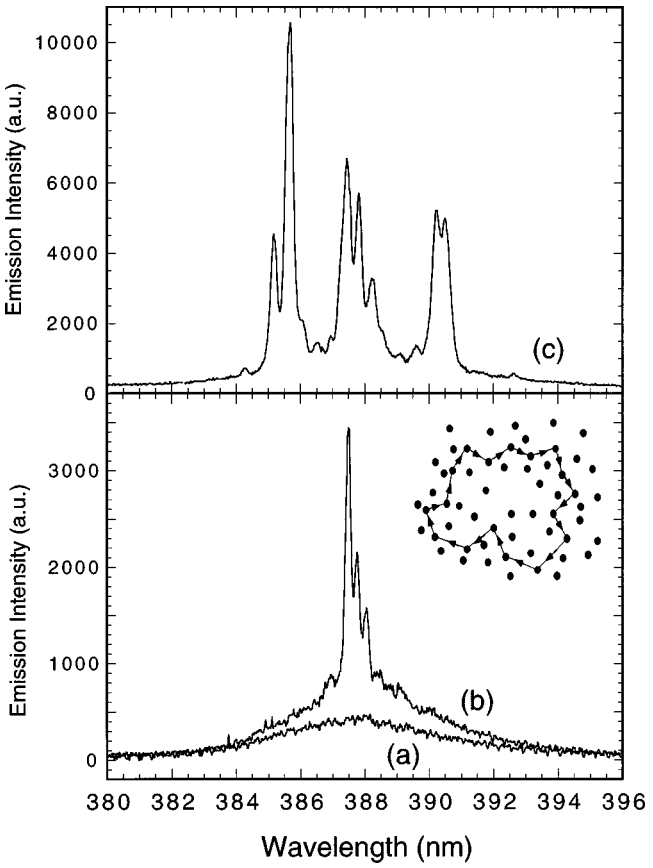

FIG. 2. Emission spectra when the excitation area is (a) 2700 μm^2, (b) 3800 μm^2, and (c) 4500 μm^2. The excitation intensity is 400 kW/cm^2. The inset is a schematic diagram showing the formation of a closed-loop path for light through multiple optical scattering in a random medium.

The next question is how were the laser cavities formed? The laser action previously observed in the ZnO films epitaxially grown on sapphire substrates occurred in the cavities formed by two cleaved facets.[5] However, we have observed lasing in the ZnO films when the excitation area (either a stripe or a spot) is far from the edges of the film (the lateral dimension of the film is about 5 mm). The dependence of the lasing threshold density on the lateral size of the excitation area rules out the possibility of lasing in the vertical cavity formed by the film surface and film–substrate interface.

Transmission electron microscopy (TEM) analysis indicates that the ZnO films have highly disordered structure. The polycrystalline grain structure of ZnO is quite evident in the plane-view TEM image. The grains appear in irregular shapes, and their sizes vary from 50 to 150 nm. This is in contrast to the well-aligned hexagonal grains in the ZnO films epitaxially grown on sapphire substrates.[6,7] The electron diffraction pattern exhibits a series of diffraction rings confirming the absence of any preferential in-plane orientation. A cross-section TEM image illustrates the columnar growth of the films. Sharp interfaces are clearly visible between adjacent columnar grains. High-resolution TEM images indicate that various types of defects, such as threading dislocation and stacking fault, are populated throughout grains. The dislocation density is estimated to be about 10^{12} cm^{-2} without including the dislocations segregated at the grain boundaries.

The in-plane randomly oriented polycrystalline grain structure of the ZnO films results in strong optical scattering. We have characterized the scattering mean-free path in the ZnO polycrystalline films using coherent backscattering.[8–10] To avoid absorption, the frequency-doubled output (410 nm) of a mode-locked Ti:sapphire laser (76 MHz repetition rate, 200 fs pulse width) was used as the probe light. From the angular width of the backscattering cone, we estimated that the scattering mean-free path is on the order of the emission wavelength of ZnO. Due to the short scattering mean-free path, the emitted light may return to the scatterer from which it was scattered before, thereby forming a closed-loop path, as shown schematically in the inset of Fig. 2.[11,12] There are many such closed-loop paths for light in the ZnO film. These loops could serve as ring cavities for light. Along different loops, the probability of a photon scattered back to its starting point is different. In other words, the ring cavities formed by scattering have different loss.

The ZnO polycrystalline films exhibit large optical gain. The gain coefficient was measured to be over 100 cm^{-1} at a

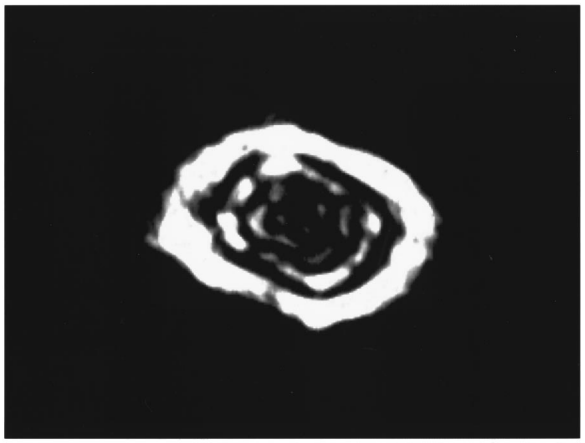

FIG. 3. Amplified images of the excitation area above the lasing threshold at different locations on the film. The excitation spot is ~35 μm in diameter and is located in the middle of the frame.

fluence of 2 μJ/cm^2. When the pump power increases, the gain exceeds the loss first in the low-loss cavities, leading to laser oscillation in these cavities. 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 observed in Fig. 1. As the pump power increases further, the gain increases and it exceeds the loss in the lossier cavities. Laser oscillation in those resonators add more discrete peaks to the emission spectrum.

Since different laser cavities could have different output directions, lasing modes observed at different angles may be different. However, some cavities may have strong output into several directions, and thus, the lasing modes from these cavities could be observed in several directions.

When the excitation area increases, there are more closed-loop paths for light, and thus, the number of lasing modes increases. On the other hand, when the excitation area is reduced to below a critical size, laser oscillation stops because the closed-loop paths are too short, and the amplification along the loops is not high enough to achieve lasing. That is why the pump intensity required to reach lasing threshold increases with a decrease of the excitation area.

To actually observe the laser resonators in the ZnO films, we have taken amplified images of the excitation area using a microscope objective (50×) and a UV sensitive charge-coupled device (CCD) camera. Below the lasing threshold we could not observe any patterns. However, above the lasing threshold, we observed the closed loops along which laser action occurred, as shown in Fig. 3. Since the lateral dimension of the excitation area is much larger than the film thickness, laser cavities are in the plane of the film. The shape and size of the two-dimensional laser cavities changed as we moved the excitation spot across the film. Simultaneous measurement of emission spectra showed that lasing mode also changed with sample position. The size and shape of the laser resonators are determined by the optical gain coefficient, the grain size and distribution, the scattering cross section, the boundary conditions, etc. Hence, the lasing frequencies vary across the film. By introducing external feedback or injection seeding, we may be able to control the lasing frequencies, and even achieve single-mode lasing.[13] Very recently, we observed similar lasing phenomena in ZnO and GaN powder with an average particle size of 100 nm. This observation provides additional support to the formation of laser resonators through multiple optical scattering.

In conclusion, we have observed self-formation of laser cavities due to strong optical scattering in semiconductor polycrystalline films. Although there have been much interest and study on lasing in highly disordered media,[14–17] to the best of our knowledge, this is the first time that the discrete cavity modes of random lasers have been observed, and actual images of two-dimensional random laser cavities have been captured. The demonstration of random lasers opens up the possibility of using disordered semiconductor microstructures as alternative sources of coherent light emission. From a practical point of view, the realization of lasing in semiconductor polycrystalline films nonepitaxially grown on amorphous fused silica substrates provides a compelling starting point for the fabrication of semiconductor lasers on many different types of substrates.

[1] N. M. Lawandy, R. M. Balachandran, A. S. L. Gomes, and E. Sauvain, Nature (London) **368**, 436 (1994).
[2] D. S. Wiersma, P. Bartolini, A. Lagendijk, and R. Righini, Nature (London) **390**, 671 (1997).
[3] H. C. Ong, J. Y. Dai, and R. P. H. Chang (unpublished).
[4] V. Srikant and D. R. Clarke, J. Appl. Phys. **81**, 6537 (1997).
[5] D. M. Bagnall, Y. F. Chen, Z. Zhu, T. Yao, S. Koyama, M. Y. Shen, and T. Goto, Appl. Phys. Lett. **70**, 2230 (1997).
[6] P. Zu, Z. K. Tang, G. K. L. Wong, M. Kawasaki, A. Ohtomo, H. Koinuma, and Y. Segawa, Solid State Commun. **103**, 459 (1997).
[7] T. Ohnishi, A. Ohtomo, M. Kawasaki, K. Takahashi, M. Yoshimoto, and H. Koinuma, Appl. Phys. Lett. **72**, 824 (1998).
[8] P.-E. Wolf and G. Maret, Phys. Rev. Lett. **55**, 2696 (1985).
[9] M. P. van Albada and A. Lagendijk, Phys. Rev. Lett. **55**, 2692 (1985).
[10] P. C. de Oliveira, A. E. Perkins, and N. M. Lawandy, Opt. Lett. **21**, 1685 (1996).
[11] Z.-Q. Zhang, Phys. Rev. B **52**, 7960 (1995).
[12] D. Wiersma and A. Lagendijk, Phys. Rev. E **54**, 4256 (1996).
[13] R. M. Balachandran, A. E. Perkins, and N. M. Lawandy, Opt. Lett. **21**, 650 (1996).
[14] V. S. Letokhov, Sov. Phys. JETP **26**, 835 (1968).
[15] S. John and G. Pang, Phys. Rev. A **54**, 3642 (1996).
[16] R. M. Balachandran and N. M. Lawandy, Opt. Lett. **22**, 319 (1997).
[17] G. A. Berger, M. Kempe, and A. Z. Genack, Phys. Rev. E **56**, 6118 (1997).