Zinc oxide thin-film random lasers on silicon substrate

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Room-temperature ultraviolet lasing is demonstrated in mirrorless zinc oxide thin-film waveguides on (100) silicon substrate. Laser cavities, due to closed-loop optical scattering from the lateral facets of the irregular zinc oxide grains, are generated through the post-growth annealing of high-crystal-quality zinc oxide thin films obtained from the filtered cathodic vacuum arc technique. It is found that the lasing wavelength and linewidth of the zinc oxide random lasers under 355 nm optical excitation are around 390 nm and less than 0.4 nm, respectively. In addition, the lasing threshold characteristics are in good agreement with the random laser theory. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1719279]

Extensive studies have been concentrated on the realization of zinc oxide (ZnO) ultraviolet (UV) lasers so that high-efficiency lasing sources can be developed.[1–4] ZnO thin-film lasers have been fabricated by plasma-enhanced molecular beam epitaxy (MBE) using cleaved facets to sustain lasing under optical excitation.[2] However, due to the natural geometry of ZnO crystals, cleaved facets are very difficult to produce. On the other hand, natural Fabry-Perot cavities inside ZnO thin films can be formed on (0001) sapphire substrate by self-assembled hexagonal grains using laser MBE at 500 °C.[1] However, the formation of self-assembled hexagonal grains can only be obtained on sapphire, which is an expensive material and difficult to process. In order to fabricate low-cost UV lasing sources and to integrate them with silicon-based electronics, it is necessary to develop a simple technique for the realization of reproducible laser cavities in ZnO thin films on silicon substrates.

Recently, we have demonstrated high-intensity amplified spontaneous emission (ASE) from ZnO thin-film waveguides deposited on silicon substrate by using the filtered cathodic vacuum arc (FCVA) technique at low temperature.[5] However, the cleaved facets of the thin-film waveguides are too rough to provide sufficient optical feedback for sustaining UV lasing. On the other hand, UV lasing has been observed in mirrorless ZnO thin films with highly disordered gain media due to the self-formation of laser cavities.[3,4] This implies that if highly disordered gain media can be realized in our thin films, it is possible to fabricate mirrorless UV lasers on lattice-mismatched silicon substrate. In this letter, the method of post-growth annealing is proposed to generate a highly disordered structure inside the ZnO thin-film waveguides for the realization of laser cavities.

The design and fabrication of ZnO thin-film waveguides have been discussed in Ref. 5 and are summarized below. Prior to the deposition of ZnO thin films, a silicon dioxide (SiO_2) buffer layer of thickness around 420 nm was formed on the *n*-type (100) silicon substrate by thermal dry oxidation. This is required because the direct deposition of ZnO on silicon will not allow the transverse confinement of light inside the ZnO thin films. Then, a 200 nm thick ZnO film was deposited on the surface of the SiO_2 buffer layer by using the FCVA technique. During the deposition, substrate temperature and oxygen partial pressure were set to 230 °C and 2×10^{-4} Torr, respectively.[6]

Low temperature deposition of ZnO thin films on the smooth surface (i.e., no preferred lateral growth direction) of amorphous SiO_2 buffer layer,[3,4] which allows the effective growth of ZnO grains and voids through post-growth annealing, is the key to realize highly disordered gain media. The growth of ZnO grains (voids) inside the ZnO thin films can be explained as the merger (evaporation) of weakly bonded ZnO grains.[7] Due to the creation of voids, the ZnO grains become loosely in contact and the corresponding lateral facets are widely exposed. Hence, the lateral facets of the irregular ZnO grains or scatterers can provide a strong optical scattering that forms closed-loop paths of lights (i.e., laser cavities). The main advantage of using post-growth annealing to form laser cavities is the controllability of the generation of scatterers through the manipulation of annealing temperature and time.

Post-growth annealing in open air was carried out in a standard Lindberg type furnace using a quartz tube reactor. Initially, the samples inside the furnace were heated up from room temperature to 900 °C within 20 min. Then, the samples were annealed for a period of time, T_a, at a constant temperature of 900 °C. Finally, the samples were cooled down to room temperature inside the furnace. This annealing process was used to minimize the buildup of thermal strain/stress at the interfaces of Si/SiO_2 and SiO_2/ZnO. It must be noted that the post-growth annealing temperature should be higher than that of the growth temperature in order to achieve effective formation of irregular ZnO grains. Annealing temperature of 900 °C was used in the process because scatterers could be generated effectively at this temperature. Figure 1 shows the scanning electron microscope (SEM) images and x-ray diffraction patterns of the samples before and after annealing. The ZnO grains of the as-grown sample are very small and closely packed. For the sample with T_a $=1$ min, it is observed that the ZnO grains become loosely

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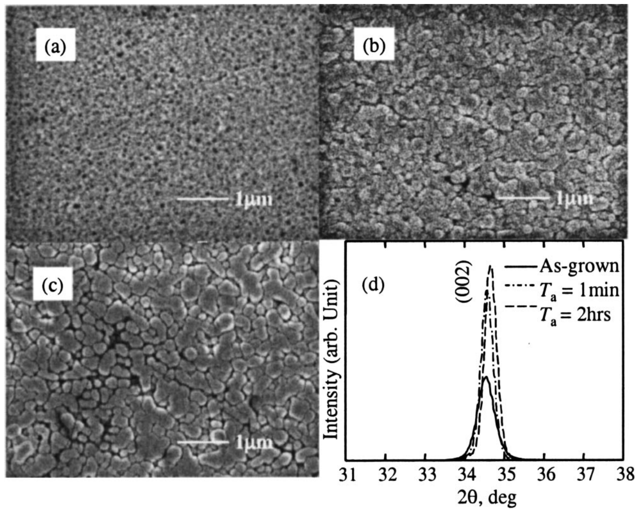

FIG. 1. SEM images of ZnO thin films: (a) as-grown, (b) $T_a = 1$ min, and (c) $T_a = 2$ h; (d) corresponding x-ray diffraction patterns.

in contact with deep slits existing between the grains (i.e., the lateral facets of the ZnO grains are beginning to develop). For $T_a = 2$ h, the size of ZnO grains grow further and almost all the ZnO grains have developed lateral facets. The corresponding x-ray diffraction patterns ($\theta/2\theta$) show a strong (002) peak at around 34.4°. This indicates that the ZnO thin films have a hexagonal wurtzite structure with their c-axis normal to the substrate-basal plane. The reduction of full width at half maximum (FWHM) of the (002) peaks implies the increase of the average size of ZnO grains after post-growth annealing, which is consistent with the observation from the SEM images. Using the method of coherent backscattering,[8] it can be shown that only the annealed samples exhibited scattering mean-free path of 390 nm which is similar to the emission wavelength of the ZnO thin films. Hence, the formation of irregular ZnO grains can create lateral facets (i.e., laser cavities) so that UV lasing is possible in the ZnO thin-film waveguides.

After the formation of highly disordered ZnO thin films, ridges with height, width, and separation of 100 nm, 2 μm, and 500 μm, respectively, were realized on the surface of the ZnO thin films by plasma etching.[5] Room-temperature optical characteristics of the ZnO thin-film ridge waveguides after post-growth annealing were studied under optical excitation by a frequency-tripled Nd:YAG laser (at 355 nm) at pulsed operation (6 ns, 10 Hz). Optical pump was achieved by using a cylindrical lens to focus a pump stripe of length 5 mm and width 8 μm on to the ridge structure. A polarizer in the direction perpendicular (TE) and parallel (TM) to the thin-film surface was also used to analyze the polarization properties of the lasing light emitted from the facets of the ridge waveguides.

Figure 2 shows the light–light curves and emission spectra of the sample with $T_a = 1$ min. At low pump intensities, a single-broad spontaneous emission peak (TE polarization) with a FWHM of about 15 nm is observed. When the excitation intensity exceeds a threshold (~ 0.35 MW/cm^2), lasing modes (i.e., sharp peaks at around 390 nm) with linewidth less than 0.4 nm emerged from the emission spectra. Further increase in pump intensity increases the number of lasing TE modes as the increase in optical gain excites more

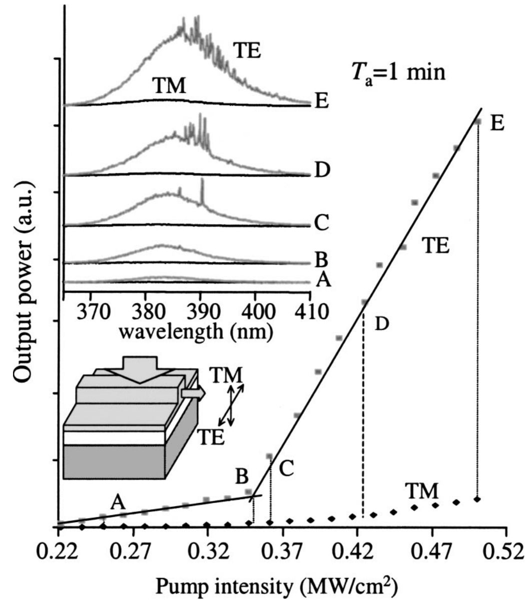

FIG. 2. Light–light curves and the emission spectra of the sample after post-growth annealing for $T_a = 1$ min.

cavity modes with higher losses. However, the lasing modes are accompanied with ASE spectra, which are similar to that of the as-grown sample.[5] This is because the laser cavities inside the ZnO thin films are weakly formed (i.e., low reflectivity of the lateral facets). On the other hand, TM polarization is strongly suppressed due to the design of the ZnO thin-film waveguides.[5]

Figure 3 shows the lasing characteristics of the sample with $T_a = 2$ h. For pump intensities exceeding 0.33 MW/cm^2, lasing modes of both polarizations are excited at around 390 nm with linewdiths less than 0.4 nm. The number of lasing modes (of both polarizations) increases with the increase of pump intensities. A second threshold is also observed at the pump intensity equal to 0.42 MW/cm^2. This is due to the excitation of the second group of laser cavities at around 400 nm with much higher cavity losses. It is observed

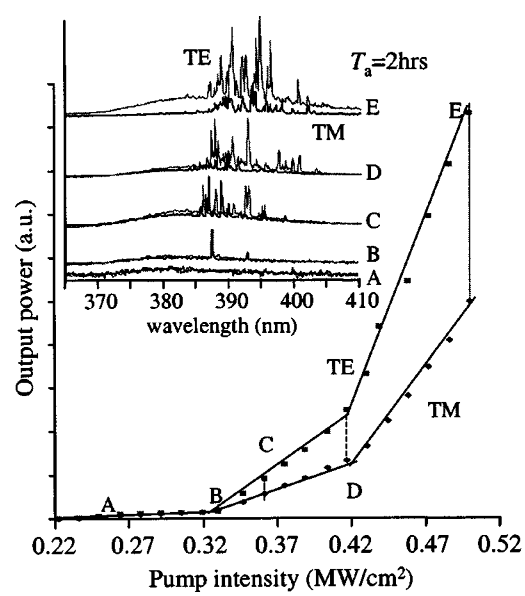

FIG. 3. Light–light curves and the emission spectra of the sample after post-growth annealing for $T_a = 2$ h.

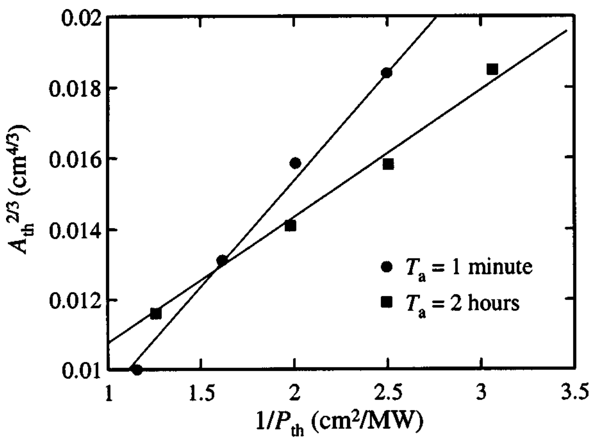

FIG. 4. Plot of $A_{th}^{2/3}$ against P_{th}^{-1} for the post-growth samples with $T_a = 1$ min and $T_a = 2$ h.

that the ASE of TE polarizations are suppressed in the lasing spectra because strong laser cavities (i.e., high reflectivity of the lateral facets) are formed inside the ZnO thin films. It must be noted that although post-growth annealing at $T_a = 2$ h will increase defect-related deep-level emission, the efficiency of near-band-edge excitonic UV emission is also increased.[7] Hence, the net optical gain of TM polarizations can be enhanced after post-growth annealing so that both TE and TM polarizations are excited simultaneously. Laser cavities can also be realized in the ZnO thin-film waveguides after an annealing temperature and time of 400 °C and 2 h, respectively. It can be shown that the lasing spectra similar to that of Fig. 2 can also be obtained in such a low annealing temperature.

Random laser theory has indicated that the critical volume $V = A_{th}$ (threshold excitation area)$\times d$(thickness of ZnO) can be related to the threshold gain length l_g and the scattering mean-free path l_s of the highly disordered gain media by $V \sim (l_s l_g)^{3/2}$.[8] If l_g is assumed to be inversely proportional to the threshold pump intensity, P_{th}, that is $l_g^{-1} = (\partial l_g^{-1}/\partial P) P_{th}^5$ and $\partial l_g^{-1}/\partial P$ is less dependent on the post-growth annealing (i.e., variation of threshold pump intensities is less than 6%), it is possible to rewrite the above equation as $A_{th}^{2/3} \sim d^{-2/3} l_s (\partial l_g^{-1}/\partial P)^{-1} P_{th}^{-1}$. Hence, the relative value of l_s of the annealed samples can be estimated by plotting $A_{th}^{2/3}$ against P_{th}^{-1} for the TE polarization. Figure 4 shows the plot of $A_{th}^{2/3}$ vs P_{th}^{-1} for both post-growth samples. It is observed in the case with $T_a = 2$ h that the corresponding slope of the fitted line is smaller than that for the case with $T_a = 1$ min. This implies that the sample with $T_a = 2$ h has shorter l_s (i.e., the lateral facets have stronger scattering strength/reflectivity) than that with $T_a = 1$ min. Hence, the threshold characteristics of the post-growth samples are in good agreement to the random laser theory.

In conclusion, post-growth annealing is proposed to generate a highly disordered structure inside the as-grown ZnO thin films. Closed-loop scattering and coherent amplification of lights are realized by highly disordered ZnO structure. Hence, high-intensity UV lasing is observed from the mirrorless ZnO thin-film waveguides on lattice-mismatched silicon substrate. It is noted that the highly disordered gain media can only be generated effectively by post-growth annealing if the ZnO thin films are deposited on SiO_2 buffer layer at low temperature. Therefore, our method to fabricate mirrorless UV lasers on silicon substrate is unique and this will make the mass production of low-cost UV lasing sources and their integration with silicon-based electronics feasible.

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