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Directional and controllable edge-emitting ZnO ultraviolet random laser diodes

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Room-temperature ultraviolet random lasing action is demonstrated from a p-GaN/annealed \( i\)-ZnO:Al(3%)/\( n\)-ZnO:Al(5%) buried heterojunction diode with a 2 \( \mu \)m rib waveguide. Excellent electrical-to-optical conversion efficiency is achieved by strong electrical and optical confinement of a buried heterojunction rib waveguide structure. Hence, emission intensity (threshold current) can be enhanced (reduced) by \( \sim 9 \) times (\( \sim 40\% \)). Directional emission as well as controllability on the number of the random lasing modes can also be achieved. © 2010 American Institute of Physics.

ZnO random media have been considered as laser cavities to fabricate electrically pumped ultraviolet (UV) laser diodes.\(^{1,2}\) This is because the difficulty to realize cleaved facets from the ZnO films can be avoided. However, high scattering loss of random media requires high optical gain to sustain random lasing action. Hence, the use of ZnO-SiO\(_2\) nanocomposite film, which has high electrical-to-optical conversion efficiency, was proposed to achieve random laser diodes.\(^{3,4}\) Alternatively, electrically pumped random laser was obtained from metal-SiO\(_2\)-polycrystalline ZnO film where the ZnO/SiO\(_2\) interface forms an electron accumulation layer to improve radiative recombination efficiency of the polycrystalline ZnO film.\(^{5}\) Similarly, a MgO thin film was used as an electron blocking barrier to reduce threshold current of the polycrystalline ZnO-GaN heterojunction laser diode.\(^{6}\) Nevertheless, poor directionality and controllability of the random lasing modes are still impairing the usefulness of the ZnO random laser diodes. In this letter, we proposed a p-GaN/annealed \( i\)-ZnO:Al(3%)/\( n\)-ZnO:Al(5%) buried heterojunction rib waveguide laser to control the excitation of random lasing modes. It can be shown that the corresponding electrical-to-optical conversion efficiency can be significantly improved. In addition, emission direction and number of random lasing modes can be controlled by the presence of the rib waveguide.

Figure 1 shows a schematic of the proposed buried heterojunction rib waveguide laser. A p-GaN:Mg/sapphire substrate with hole concentration of \( \sim 5 \times 10^{17} \) cm\(^{-3}\) was used as the hole injection layer and substrate. A \( \sim 150 \) nm thick ZnO:Al(3%) thin film was deposited onto half of the p-GaN:Mg/sapphire substrate by filtered cathodic vacuum arc (FCVA) technique. During the deposition, substrate temperature and oxygen partial pressure were set to \( \sim 150 \) °C and \( \sim 2 \times 10^{-3} \) Torr, respectively.\(^7\) The sample was annealed at 900 °C for 30 min in open air to form highly disordered ZnO grains and voids in order to sustain random lasing action. In addition, the use of Al-doped ZnO film as the rib waveguide is to maintain electrically conductive after the annealing process. The annealed \( i\)-ZnO:Al(3%) is found to have electron concentration and mobility of \( \sim 5 \times 10^{16} \) cm\(^{-3}\) and \( \sim 6 \) cm\(^2\) V\(^{-1}\) s\(^{-1}\), respectively. Subsequently, a line-mask (with width, thickness, and separation equal to 2, 0.8, and 500 \( \mu \)m, respectively) was coated onto the surface of the annealed \( i\)-ZnO:Al(3%) film by photolithography technique. The unmasked \( i\)-ZnO:Al(3%) layer was then completely removed by ion-beam sputtering with an etching rate of \( \sim 10 \) nm/min for 15 min.\(^8\) The inset of Fig. 2 shows a scanning electron microscope (SEM) image of the \( i\)-ZnO:Al(3%) rib waveguide after postgrowth annealing and ion-beam sputtering. Highly disordered ZnO grains are clearly observed from the SEM image.

A \( \sim 120 \) nm thick of SiO\(_2\) cladding layer was then deposited onto the sample by E-beam evaporation with substrate temperature set to 50 °C. After the deposition, a lift-off process was carried out to remove the excess SiO\(_2\) layer attached onto the surface of the annealed \( i\)-ZnO:Al(3%) rib waveguides. The SiO\(_2\) cladding layer was used as an electrical isolation layer to prevent the lateral diffusion of injection carriers from the rib waveguide.\(^9\) As the refractive index of SiO\(_2\) cladding layer (\( n = 1.45 \)) is smaller than that of the \( i\)-ZnO:Al(3%) rib (\( n = 2.1 \)), strong lateral optical confinement can also be achieved. Finally, a layer of \( n\)-ZnO:Al(5%) with thickness of \( \sim 150 \) nm was deposited onto the surface of the sample by the FCVA technique to serve as an electron injection layer. The deposition conditions are the same as that of the ZnO:Al(3%) layer. The carrier concentration of \( n\)-ZnO:Al(5%) film was found to be \( \sim 10^{21} \) cm\(^{-3}\). A \( \sim 100 \) nm thick Au (Ni) films were deposited onto the p-GaN:Mg/sapphire substrate (\( n\)-ZnO:Al(5%) layer) as the p-type (n-type) metal contact by E-beam evaporation. For the purpose of comparison, another p-GaN/annealed \( i\)-ZnO:Al(3%)/\( n\)-ZnO:Al(5%) buried heterojunction rib waveguide laser.
$i$-ZnO:Al(3%)$/n$-ZnO:Al(5%) heterojunction laser diode without a rib waveguide structure was also fabricated.

Figure 2 also compares the photoluminescence (PL) spectra of the as-grown and annealed ZnO:Al(3%) layer deposited on GaN:Mg/sapphire substrate as well as a bare GaN:Mg/sapphire substrate under the excitation by a 355 nm frequency tripled neodymium-doped yttrium aluminum garnet pulsed laser (10 Hz, 6 ns). It is observed that the as-grown ZnO:Al(3%) exhibits a broad spontaneous emission with peak located at $\sim$385 nm. After annealing at 900 °C in open air for 30 min, lasing modes (i.e., sharp peaks) at $\sim$385 nm are emerged from the emission spectrum. This is because highly disordered ZnO grains and voids are generated from the ZnO:Al(3%) by thermal annealing. The size of the ZnO grains, which have an average diameter of $\sim$100 nm, are sufficiently large to support coherent random lasing. On the other hand, the GaN:Mg layer shows an emission peak at $\sim$367 nm.

In order to demonstrate the importance of the proposed buried heterojunction rib waveguide structure, lasing characteristics of the ZnO random laser diodes with and without a buried rib waveguide are compared. Figure 3(a) shows the edge and surface emission spectra of the ZnO random laser diode without a buried rib waveguide under various forward biases. For the injection current, $I$, $\geq$5 mA, sharp peaks are emerged from the edge emission spectra at around 385 nm. In addition, the number of sharp peaks increases with the increase in $I$. This is attributed to the coherent random lasing action inside the annealed $i$-ZnO:Al(3%) layer. However, only spontaneous emission is observed from the surface of the laser diode. For $I$\approx8.3 mA, the emission peaks observed from the edge and surface of the laser diode are blueshifted to 375 and 370 nm, respectively, which are closed to the emission peak of GaN:Mg layer. This indicates that the injected electrons are drifted to the $p$-GaN:Mg layer and radiative recombination was taken place at the interface between the $p$-GaN:Mg and $i$-ZnO:Al(3%) layers. This implies that the transverse electrical confinement of the $p$-$i$-$n$ heterojunction is deteriorated by the large value of $I$ due to current crowding effect.

Figure 3(b) shows the corresponding light-current ($L$-$I$) curves of the ZnO random laser diode without a buried rib waveguide. It is observed that there is an obvious kink at $I$ \sim 5.0 mA in the $L$-$I$ curve for the case of edge emission. This corresponds to the threshold current for the lasing modes emitted from the edge of the laser diode. Furthermore, the intensity of surface emission is less than that of edge emission. This indicates that the random lasing action is confined within the highly disordered $i$-ZnO:Al(3%) layer and the scattering of light from the surface and bottom of the annealed $i$-ZnO:Al(3%) layer is suppressed by the relatively low refractive indices of electron and hole injection layers via total internal reflection. The near field image of the diode laser, which verifies the transverse optical confinement of the laser diode, is also shown in the inset of Fig. 3(b). Nevertheless, lateral confinement of the laser diode has not been achieved.

Edge emission spectra of the diode laser with a buried rib waveguide are plotted in Fig. 4(a). For $I$\approx3.2 mA, sharp peaks at around 387 nm are emerged from the edge emission spectrum. For further increase in $I$, the number and intensity of the lasing peaks are also increased. However, the number of dominant sharp peaks is less than that shown in Fig. 3(a). The corresponding surface emission spectra, which exhibit only broad spontaneous emission, are also plotted in the inset of Fig. 4(a). From the corresponding $L$-$I$ curves shown in Fig. 4(b), it is noted that the intensity of edge emission (threshold current) is enhanced by $\sim$9 times (reduced from 5 to 3.2 mA) when compared to that without the buried rib waveguide. Furthermore, at a large value of $I$, the lasing peaks were excited electrically at around 387 nm which is
closed to the emission peak of ZnO:Al(3%) film. This implies that the electrical confinement of the $p-i-n$ heterojunction inside the annealed $i$-ZnO:Al(3%) waveguide is improved at large value of $I$ and the influence of current crowding is suppressed. Hence, the electrical-to-optical conversion efficiency of the laser diode is strongly enhanced by the presence of the 2 $\mu$m rib waveguide embedded inside the SiO$_2$ cladding layer. The near field image of the edge emission is also shown in the inset of Fig. 4(a). Only a single light spot is observed from the edge of the rib waveguide. Hence, these indicate that strong lateral and transverse optical confinement can be achieved simultaneously from the rib waveguide structure and the edge emission is highly directional.

If the lasing mechanism of the laser diode is related to coherent random laser action, it is possible to deduce the closed-loop cavity length, $L$, of the corresponding random modes by power Fourier transform (FT).\textsuperscript{11} The inset of Fig. 4(b) shows the power FT of the laser diodes with and without a buried rib waveguide biased at two times of its threshold current. It is found the presence of rib waveguide reduces the value of $L$ from $\sim$6.8 to 2.3 $\mu$m. This implies that the size of the closed-loop random cavities that can be formed inside the annealed $i$-ZnO:Al(3%) layer is limited by the width of the rib waveguide. As spatial overlapping of closed-loop cavity modes is not allowed inside a random medium due to the repulsion mechanism of lasing modes,\textsuperscript{12} the number of excited lasing modes will be limited. This explains why there are much fewer dominant lasing peaks observed from the emission spectra of the buried heterojunction rib waveguide laser.

In summary, coherent random lasing action is realized inside the annealed $i$-ZnO:Al(3%) rib waveguide due to the formation of ZnO grains and voids. Relatively low resistivity of $i$-ZnO:Al(3%) layer allows the effective injection of carriers through the $n$-ZnO:Al(5%) electron injection layer and $p$-GaN hole injection layer. In addition, the injection efficiency of carriers into the annealed $i$-ZnO:Al(3%) rib waveguide can be further improved by suppressing the lateral diffusion of carriers through the use of SiO$_2$ cladding layer as an electrical isolation layer. On the other hand, scattering loss of the annealed $i$-ZnO:Al(3%) rib waveguide is significantly reduced via total internal reflection from the electron and hole injection layers as well as the SiO$_2$ cladding layer. Hence, the $p-i-n$ buried heterojunction rib waveguide structure has maximized the electrical and optical confinement along the lateral and transverse directions of the laser diode. Furthermore, highly collimated emission beam is observed from the edge of the laser diode due to the strong optical confinement of the buried rib waveguide. As the size of closed-loop cavity modes is limited by the width of $i$-ZnO:Al(3%) rib waveguide, the number of lasing peaks that can be excited will also be restricted. Therefore, we have verified that the proposed UV emission $p$-GaN/annealed $i$-ZnO:Al(3%)/$n$-ZnO:Al(5%) buried heterojunction rib waveguide laser diode has high electrical-to-optical efficiency. The corresponding emission beam is directional, and the number of random lasing modes can be controlled by the presence of rib waveguide structure.

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