JOURNAL OF APPLIED PHYSICS **106**, 043102 (2009)

High-temperature lasing characteristics of randomly assembled ZnO nanowires with a ridge waveguide

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(Received 8 May 2009; accepted 9 July 2009; published online 19 August 2009)

High-temperature lasing characteristics of randomly assembled ZnO nanowires at ultraviolet wavelength are investigated. Lasing emission was observed from the randomly assembled ZnO nanowires for temperature up to 500 K. The corresponding lasing mechanism is attributed to the coherent random lasing action via the formation of closed-loop cavity modes. Furthermore, ridge waveguide lasers using the randomly assembled ZnO nanowires as the active medium were fabricated. It is found that the width of the ridge waveguides restricted the size of the closed-loop cavity modes such that the corresponding characteristic temperature can be increased by 10 K.
 [DOI: 10.1063/1.3200960]

I. INTRODUCTION

The use of random media such as cluster of ZnO nanopowder,[1] randomly assembled ZnO nanowires,[2,3] and highly disordered ZnO thin films[4] as the active layer of ultraviolet (UV) semiconductor lasers has been proposed to avoid the formation of smooth facets. This is possible because the highly scattering random media allow the formation of closed-loop cavity modes so that optical feedback is not required from the facets.[5] However, the main drawback of the closed-loop cavity modes is that their orientation inside the random media is less controllable.[6] As a result, different closed-loop cavity modes will exhibit different cavity losses and lasing directions. Furthermore, high-temperature performance of the random media can be deteriorated due to the weak optical confinement of the closed-loop cavity modes.[7] Hence, it is highly desired to control the orientation of the closed-loop cavity modes in order to improve the lasing performance of the random media.

Optical waveguide is an important transmission medium to confine and control the propagation of light.[8] This method has also been extended to enhance the lasing performance of random media.[6] In this paper, the possibility to improve the high-temperature lasing characteristics of the randomly assembled ZnO nanowires by using optical waveguide is investigated. Ridge waveguide geometry is proposed to control the size of the closed-loop cavity modes formed inside the randomly assembled ZnO nanowires. It can be shown that the lasing threshold can be reduced and the emission beam can be collimated. The high-temperature performance of the random media can also be enhanced. It is found that the improvement of high-temperature performance is dependent on the width of the optical waveguide which is related to the diameter of the closed-loop cavity modes.

II. FABRICATION AND CHARACTERIZATION OF RANDOMLY ASSEMBLED ZNO NANOWIRES

Randomly assembled ZnO nanowires were grown by vapor transport method inside a horizontal tube furnace.[9] A (111)-oriented Si substrate, which has a 1 μm thick SiO_2 buffer layer, was used as a template to grow the ZnO nanowires. Before the fabrication, the Si substrate was ultrasonically cleaned in acetone and then coated with a 2 nm thick Au catalyst film onto the SiO_2 buffer layer. The source, which consists of 0.1 g of ZnO:C powder mixture with a weight ratio of 3:2, was loaded into a small quartz tube with an inner diameter of 1.4 cm. The Au coated Si substrate was placed 2 cm downstream from the source. The quartz tube was inserted into a larger quartz tube mounted on the furnace. It is noted that the source was positioned at a distance 5 cm downstream from the center of the furnace. The large quartz tube was first pumped down to 10^{-2} mbar and then it was flushed three times with a carrier gas (99.5% Ar, 0.5% O_2). During the fabrication, the temperature at the center of the furnace was increased at a constant rate of 50 °C/min from room temperature until it reached 960 °C. Furthermore, a constant flow of the carrier gas at 50 SCCM (SCCM denotes cubic centimeter per minute at STP) was maintained and pressure was approximately kept at 20 mbar inside the large quartz tube. After the growth of ZnO nanowires for 1 h, the furnace was cooled down to room temperature. It can be shown that a layer of 800 nm thick randomly assembled ZnO nanowires was deposited on the SiO_2 buffer layer.

Figure 1(a) shows the field emission scanning electron microscopy (SEM) image of the randomly assembled ZnO nanowires. The length of the nanowires is ranging from 1 to 5 μm and the corresponding diameter is about 100 nm. The x-ray diffraction (XRD) pattern of the ZnO nanowires, which XRD peaks are related to the wurtzite ZnO structure, is also given in Fig. 1(b). Figure 1(c) shows the transmission electron microscopy (TEM) image of a single ZnO nanowire. High-resolution TEM image and selective area electron dif-

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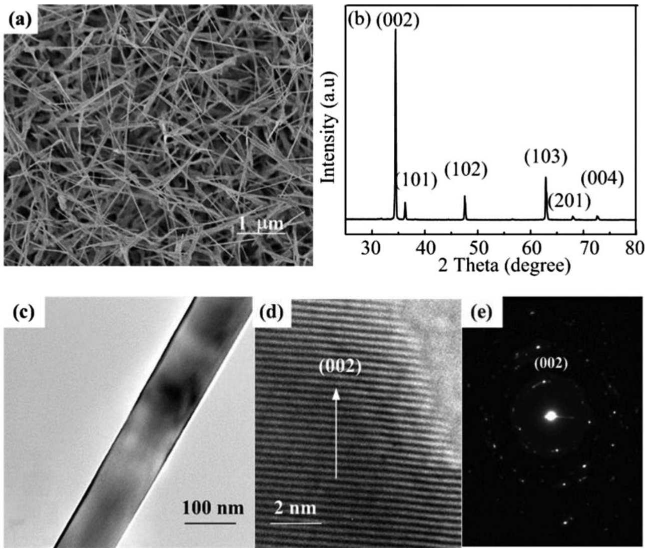

FIG. 1. (a) SEM image of the randomly assembled ZnO nanowires; (b) XRD pattern, SEM image of a ZnO nanowire; (d) TEM image of the ZnO nanowire; (e) the corresponding SAED pattern.

fraction (SAED) pattern of the ZnO nanowire are also shown in Figs. 1(d) and 1(e), respectively. The high-resolution TEM image illustrates that the ZnO nanowires have high crystal quality and wurtzite structure with lattice constants $a = 0.32$ nm and $c = 0.52$ nm.[10]

Optical characteristics of the sample were studied by using a frequency-triple 355 nm pulsed Nd:YAG (yttrium aluminum garnet) laser (120 ps pulse width and 10 Hz repetition rate) as the excitation source.[4] A cylindrical lens was used to focus a pump stripe of ~ 500 μm wide onto surface of the sample. Emission was collected by an objective lens from the edge of the sample. Figure 2 shows the evolution of emission spectra as a function of pump intensity. When the pumping intensity reached the threshold of ~ 0.35 MW/cm^2, a peak with linewidth of ~ 0.4 nm emerged from the single-broad emission spectra. As the pump power further increases, more shape peaks are excited from the emission spectrum between 380 and 390 nm. The inset of Fig. 2 shows the far field profile observed from the edge of the sample. It is noted that the emission light is TE dominated. This is because the randomly assembled ZnO nanowires appear like a slab waveguide to the lasing modes so that TE polarization is more likely to be supported. In addition, the emission beam is less collimated. The emission spectra and light-light curve of the randomly assembled ZnO nanowires have verified that the sample exhibits lasing characteristics and the corresponding lasing mechanism can be attributed to random lasing action.[2,7] The lasing mechanism of the randomly assembled ZnO nanowires will be further investigated at the last paragraph of this section.

Lasing characteristics of the randomly assembled ZnO nanowires at high temperature are also investigated. This can be done by mounting the sample onto an electrical ceramic heater of size 1×1 cm^2. Hence, temperature of the sample is allowed to vary between 300 and 500 K. Figure 3(a) shows the emission spectra of the sample versus temperature, T. The sample was pumped at about $2 \times P_{th}$, where P_{th} is the excitation threshold intensity and its value increases with the increase of T. It is noted that lasing peaks can only be excited from the sample up to $T = 500$ K. Figure 4(b) plots the domi-

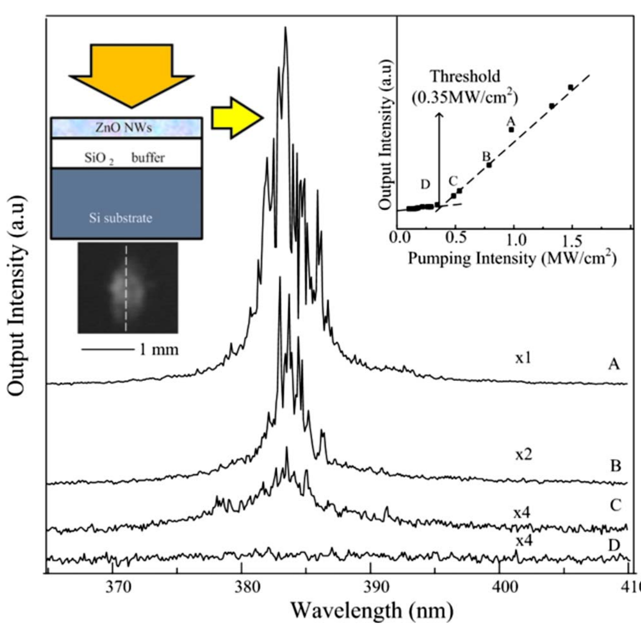

FIG. 2. (Color online) Lasing spectra and light-light curve of the randomly assembled ZnO nanowires at room temperature. The inset shows the corresponding far field profile at pump intensity of $2 \times P_{th}$. The dashed line indicates the location of the sample.

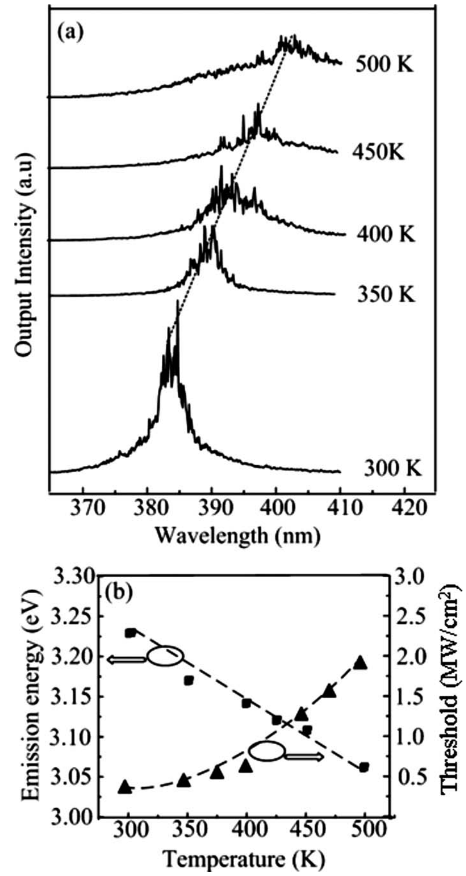

FIG. 3. (a) Lasing spectra of the randomly assembled ZnO nanowires vs T. The sample was pumped at $2 \times P_{th}$. (b) Plot of lasing peak energies and P_{th} of the sample vs T.

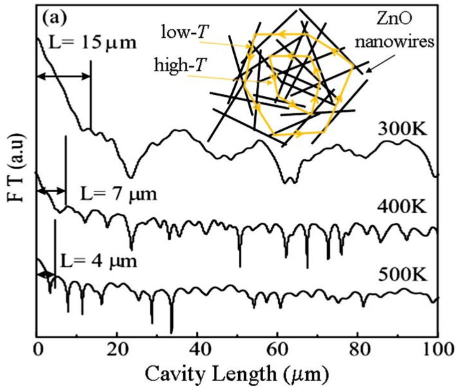

FIG. 4. (Color online) Plot of FT of the lasing spectra of the randomly assembled ZnO nanowires operating at 300, 400, and 500 K and pumped at $2\times P_{\mathrm{th}}$. The arrow indicates the cavity length of the fundamental closed-loop cavity modes.

nant peak energies and P_{th} of the randomly assembled ZnO nanowires as a function of T. The energy of the dominant peaks redshifts with the increase in T and at a constant rate of -0.7 meV/K for the entire range of T. It is observed that the emission peak energies and T has a linear relationship. This implies that the redshift of lasing peaks is mainly due to the shrinkage of the bandgap with the increase in T.[11] The dominant lasing peaks redshifted from 3.24 to 3.1 eV indicated that the radiative recombination process of the ZnO nanowires is mainly due to exciton-exciton scattering.[12] Furthermore, the value of P_{th} increases exponential with the increase in T. The solid line represents the best fit using least-squares fitting of the experimental data to the empirical formula: $P_{\mathrm{th}}(T)=P_0 \exp(T/T_c)$, where P_0 is the threshold pump intensity at $T=0$ K and T_c is the characteristic temperature. T_c is an indicator on the temperature stability level (temperature insensitivity) of the lasers. The higher the value of T_c can be obtained, the better the high-T laser performance can be obtained.[13] From Fig. 3(b), it is found that the T_c of the random assembled ZnO nanowires is 88 K. Report has shown that temperature dependence of stimulated exciton-exciton scattering of the ZnO films on sapphire and ZnO/ZnMgO superlattices on $ScAlMgO_4$ have the values of T_c equal to 67 and 87 K, respectively.[14] Hence, the high-T performance of the randomly assembled ZnO nanowires, which may be a better choice to realize UV semiconductor lasers, is compatible to that of ZnO thin films.

If the lasing mechanism of the randomly assembled ZnO nanowires is related to coherent random laser action, it is possible to deduce the cavity length of the corresponding closed-loop cavity modes by Fourier transform (FT). Figure 4 plots the FT of the lasing spectra of the randomly assembled ZnO nanowires at T equal to 300, 400, and 500 K under an optical excitation of $2\times P_{\mathrm{th}}$. A series of broad peaks of harmonics, which corresponding to different cavity lengths of the random modes, are observed from the FT patterns. The closed-loop path length, L, of the fundamental cavity modes at T equal to 300, 400, and 500 are found to be $\sim$15, $\sim$7, and $\sim$4 μm, respectively. If the closed-loop paths of the coherent photon are approximately circles, the maximum diameters, D, of these lasing cavities will be equal to

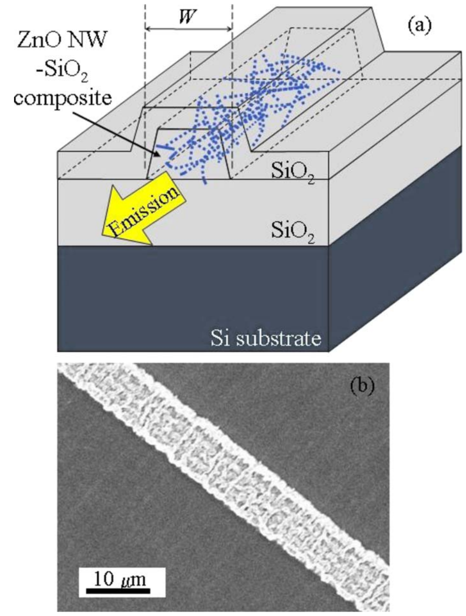

FIG. 5. (Color online) (a) Schematic of ridge waveguide lasers embedded with randomly assembled ZnO nanowires. (b) SEM image of the ZnO nanowires after etching by ion-beam sputtering. A line mask of 10 μm was used in the etching process.

$\sim$5, $\sim$2, and $\sim$1.3 μm at T of 300, 400, and 500 K, respectively. It is noted that all the estimated values of D are shorter than the width of the pump stripe and the value of D reduces with the increase in T. This is because the excitation beam has a Gaussian profile and the location of the highest gain region is at the middle of the pump stripe.[7] Furthermore, Fig. 1(a) has indicated that some regions of the randomly assembled nanowires have separation between two nanowires of less than $\sim$300 nm so that closed-loop path of D $\sim$1.3 μm can be sustained. At low T, random modes with long L can receive a high average optical gain to sustain coherent random lasing. When T increases, only the cavity modes with short L can receive a high optical gain near the middle of pump stripe. Hence, the value of L reduces with the increase in T. This self-compensation mechanism of cavity modes maintains the optical gain for lasing emissions at high T. In fact, these observations are consistent with our finding in ZnO thin films[7] so that the lasing mechanism of the randomly assembled ZnO nanowires should be related to coherent random lasing.[14]

III. BURIED RANDOMLY ASSEMBLED ZNO NANOWIRES RIDGE WAVEGUIDE LASERS

From the above studies of the lasing characteristics of the randomly assembled ZnO nanowires, it is noted that if the cavity modes can be restricted to a short L, random lasing action can be survived at a high T. Hence, it is believed that by laterally confining the cavity modes inside an optical waveguide, it is possible to improve the high-T performance of the random lasers. Figure 5(a) shows the schematic of a proposed buried ZnO nanowires ridge waveguide random laser to achieve better high-T performance. The fabrication process of the ridge waveguide lasers can be explained as follows.

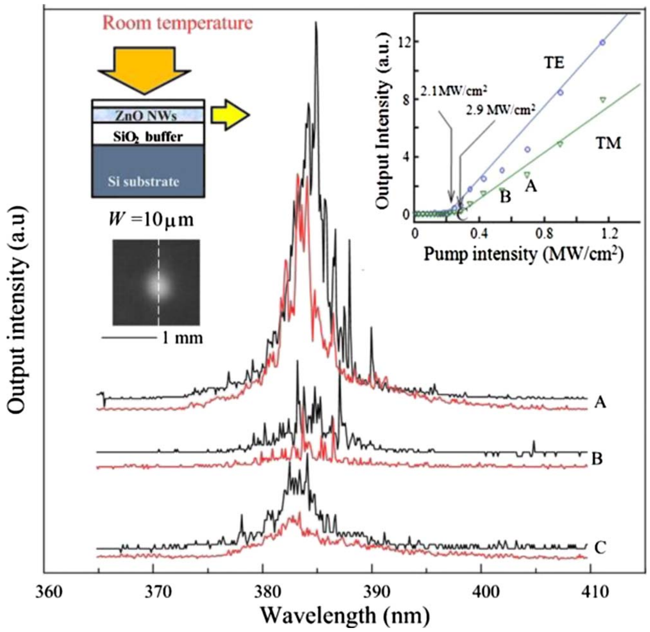

FIG. 6. (Color online) Light–light curves and emission spectra of the ridge waveguide ZnO nanowires laser with $W=10$ μm measured at room temperature. The inset shows the corresponding TE emission image far field profile at pump intensity of $2\times P_{th}$. The dashed line indicates the location of the sample.

(1) A line mask of width, W, was made on the surface of the randomly assembled ZnO nanowires by photolithography technique.

(2) The unmasked ZnO nanowires region was then etched away by Ar^+ ion-beam sputtering. The ion-beam energy and current of the Kaufmann-type ion-beam source were set to ~ 2 keV and 60 mA, respectively, so that the etching rate was ~ 30 nm/min at a chamber pressure of 3×10^{-4} Torr. The etching process was carried for more than 30 min at room temperature until the etching reached the SiO_2 buffer layer. Figure 5(b) shows the SEM image of the randomly assembled ZnO nanowires after etching by the ion-beam sputtering. It is observed that a rib structure of the randomly assembled ZnO nanowires with width of 10 μm was formed on the SiO_2 buffer layer. The randomly distribution of ZnO nanowires along the rib was clearly shown in the SEM image.

(3) After the removal of line mask, a SiO_2 capped layer of ~ 400 nm thick was deposited on to the sample by e-beam sputtering of SiO_2. Hence, a buried randomly assembled ZnO nanowires ridge waveguide laser was formed.

Figure 6 shows the room temperature lasing characteristics of the buried randomly assembled ZnO nanowires ridge waveguide laser with $W=10$ μm. It is observed that a sharp peak of TE (TM) polarization started to emerge for pump intensity reach the threshold of 0.21 MW/cm^2 (0.29 MW/cm^2). The excitation of TM polarization is due to the lateral confinement of the ridge waveguide.[8] Nevertheless, the threshold is significantly reduced (the reduction can be as much as 60% for the TE polarization at room temperature) when compared to that given in Fig. 2. This implies that

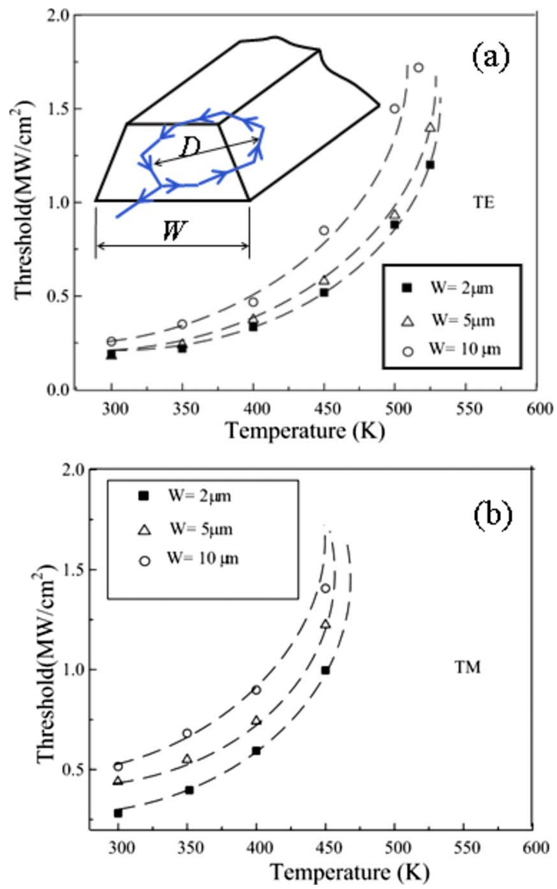

FIG. 7. (Color online) Plots of P_{th} vs T with different stripe widths ($W=2,5,10$ μm) for (a) TE and (b) TM polarizations. The inset in (a) shows the schematic of the formation of a closed-loop cavity mode inside the ridge waveguide.

the scattering loss of the randomly assembled nanowires has been reduced by the SiO_2 capped layer. The TE polarized far field profile emitted from the edge of the sample is also shown in the inset of Fig. 6. It is observed that the ridge waveguide structures exhibit a single bright-spot emission with significant reduction in beam divergence when compared to that given in Fig. 2. The improvement of lasing performance can be explained by the trapping of scattering light inside the ridge waveguide. The SiO_2 capped layer traps light from scattering away from the ZnO nanowires by total internal reflection. As a result, the ridge waveguide restricted the emission directions of the lasing beam so that beam divergence and scattering loss of the randomly assembled ZnO nanowires are reduced simultaneously.

Figure 7 plots the values of P_{th} for both TE and TM polarizations versus T of the ridge waveguide lasers with $W=10$, 5, and 2 μm. It is observed that over the entire range of T, P_{th} of both polarizations can be reduced with the decrease in W. However, P_{th} of TE polarizations is lower than that of TM polarizations. This is because the TE polarizations experienced stronger optical feedback than that of TM polarizations. On the other hand, the values of T_c of the TE (TM) polarizations were found to be increased from 92 (86) to 98 (92) K for the reduction in W from 10 to 2 μm, respectively. This implies that the presence of ridge waveguide can improve the high-T performance of the randomly assembled ZnO nanowires lasers especially for the waveguide with small W. This is because the narrow width of the ridge waveguide, which restricted the diameter, D, of the closed-loop cavity modes can be formed [see the inset of Fig. 7(a)], ensuring the effective excitation of the low-loss lasing modes. As we have shown in Fig. 4, small closed-loop cavity modes can survive at high-T; the presence of ridge waveguide will restrict the generation of lasing modes with D larger than W. At $T=300$ K, the maximum D to achieve high quantum conversion efficiency for the TE polarizations is found to be about 5 μm (equivalent to $L \sim 15$ μm, see Fig. 4); therefore further reduction in W below 5 μm should have no influence on the value of P_{th}. In fact, Fig. 7(a) has verified that P_{th} will not be further reduced for the decrease in W from 5 to 2 μm at $T=300$ K. The redshift of dominant peak energies versus T for both TE and TM polarizations is similar to that given in Fig. 3(b) so the plots are not repeated. Hence, we have verified that the use of ridge waveguide can improve the high-T performance of ZnO nanowires random lasers.

IV. CONCLUSION

The high-T lasing characteristics of randomly assembled ZnO nanowires fabricated by vapor transport method were studied. It is noted that the TE dominant lasing emission was observed from the edge of the randomly assembled ZnO nanowires film with P_{th} measured to be 0.35 to 1.9 MW/cm^2 for T varies from 300 to 500 K, respectively. However, lasing emission was not observed for T larger than 500 K. Furthermore, it is found that the corresponding lasing mechanism was due to the coherent random lasing action via the formation of closed-loop cavity modes. As a result, the randomly assembled ZnO nanowires led to less collimation of the emission beam and high scattering losses of the closed-loop cavity modes.

It is noted that the maximum available value of D (i.e., diameter of the closed-loop cavity modes) is restricted to some values and its value is dependent on the magnitude of T. This implies that if all generated closed-loop cavity modes can be limited to a particular value of D, high-T performance of the randomly assembled ZnO nanowires can be improved. This is because the excitation of high-loss closed-loop cavity modes can be avoided so that the value of P_{th} will be reduced. Therefore, it is proposed to realize a ridge waveguide laser using randomly assembled ZnO nanowires as the active medium. Due to the guiding mechanism of the optical waveguide, the maximum value of D that can be supported inside the ZnO nanowires will be restricted to the value of W. TM polarization is also excited by the optical waveguide. It is found that the value of P_{th} and the divergence of the emission beam of both TE and TM polarizations can be reduced over the range of T. Furthermore, T_c of the ridge waveguide lasing with W equal to 2 μm can be improved by 10 K for the TE polarization. Hence, we have verified that the use of waveguide geometry can be used to improve the high-T lasing performance of random media.

ACKNOWLEDGMENTS

This work was supported by the LKY PDF 2/08 startup grant. The authors (C.P. and B.Y.) are grateful for used of materials from Professor Tom Wu.

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