Random laser action in ZnO nanorod arrays embedded in ZnO epilayers

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Random laser action with coherent feedback has been observed in ZnO nanorod arrays embedded in ZnO epilayers. The sample was fabricated by depositing a MgO buffer layer and followed by a layer of ZnO thin film onto a vertically well-aligned ZnO nanorod arrays grown on sapphire substrate. Under 355 nm optical excitation at room temperature, sharp lasing peaks emit at around 390 nm with a linewidth less than 0.4 nm has been observed in all directions. In addition, the dependence of the lasing threshold intensity on the excitation area is shown in good agreement with the random laser theory. Hence, it is demonstrated that random laser action can also be supported in ZnO nanorod arrays. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1734681]

Room-temperature lasing in ZnO nanorod arrays has been demonstrated under 355 nm optical excitation with threshold pump intensity of 40 kW/cm^2.[1,2] It is believed that the optical feedback between the top and bottom natural facets of the ZnO nanorods is the lasing mechanism. On the other hand, high-density ZnO nanorod arrays can also form random lasers if the corresponding gain length and scattering mean-free path satisfy the requirement of random laser action.[3] In fact, random lasing has been observed in ZnO nanorod arrays embedded in anodic Al$_2$O$_3$ with a threshold pump intensity of 100 kW/cm^2 although the authors did not confirm the lasing mechanism is due to the random laser action.[4] In this letter, we demonstrate the possibility of achieving random laser action with coherent feedback in ZnO nanorod arrays.

ZnO nanorods were grown on sapphire at about 400 °C by using a metalorganic vapor-phase epitaxy system.[5] The ZnO nanorods are aligned normal to the surface of the sapphire substrate with an average diameter, length, and density of 70 nm, 2 μm, and 1.7×10^{11} nanorods/cm^2, respectively. The optical characteristics of the ZnO nanorods were studied at room temperature under optical excitation by a frequency tripled (355 nm) Nd:YAG laser at pulsed operation (6 ns and 10 Hz). The pump beam illuminated on the ZnO nanorods at an incident angle (varies between 0° and 80°) to the surface normal of the substrate. The emission light was collected in the directions normal as well as parallel to the substrate. Our measurement has shown that only amplified spontaneous emission (ASE) spectra with a peak wavelength and full width half maximum (FWHM) of about 380 nm and 15 nm, respectively, are observed at a pump intensity greater than 3 MW/cm^2. The threshold gain, g_{th}, of the ZnO nanorods can be estimated from $g_{th} = L^{-1} \ln(r_{sub}^{-1} r_{air}^{-1})$, where L (=2 μm) is the length of the ZnO nanorods, r_{sub} (=0.325) and r_{air} (=0.355) are the field reflectivities at the ZnO/sapphire and

ZnO/air natural facets, respectively. g_{th} is derived under the ideal Fabry–Perot (FP) cavity approximation with no internal absorption loss is taken into consideration. It is found that the calculated value of g_{th} is roughly equal to 11×10^3 cm^{-1}. If the influence of diffraction losses is taken into consideration, the actual required value of g_{th} could be doubled.[6] This implies that the required value of g_{th} to sustain lasing in the natural FP cavities of the ZnO nanorods is very large, larger than the ZnO material can provide. Hence, the natural FP cavities of the ZnO nanorod arrays will not sustain lasing even under a high optical excitation.

The above measurement has also indicated that the ZnO nanorod arrays do not support random lasing due to the low density of nanorods. In order to satisfy the requirement of random laser action, the gain length (scattering mean free path) of the ZnO nanorod arrays has to be increased (reduced).[3] This can be achieved by forming a low-loss slab waveguide, which allows strong transverse confinement of light inside the ZnO thin film, along the surface parallel to the sapphire substrate growth with ZnO nanorod arrays. Figure 1 shows the fabrication procedures of the ZnO nanorods embedded in ZnO epilayers. A MgO buffer layer of thickness 700 nm was deposited onto the ZnO nanorod arrays and followed by a ZnO film of thickness 200 nm by using filtered cathodic vacuum arc technique at 230 °C.[7] The purposes of forming the slab waveguide are: (1) To provide an extra gain length and (2) to reduce the scattering mean-free path (i.e., equivalent to minimize the average separation between the ZnO nanorods, that occurs when light only travels in the direction parallel to the substrate). It must be noted that the refractive indices of MgO and ZnO are roughly equal to 1.76 and 2.1, respectively, at 390 nm and the effective refractive index of the slab waveguide can be estimated to be 1.99.[8] Hence, light scattering formed by the ZnO nanorods is maintained which is due to the discontinuity in refractive index between the boundaries of the slab waveguide and nanorods. If the sample is under optical excitation (i.e., see also Fig. 1), optical amplification and scattering of the guided modes can

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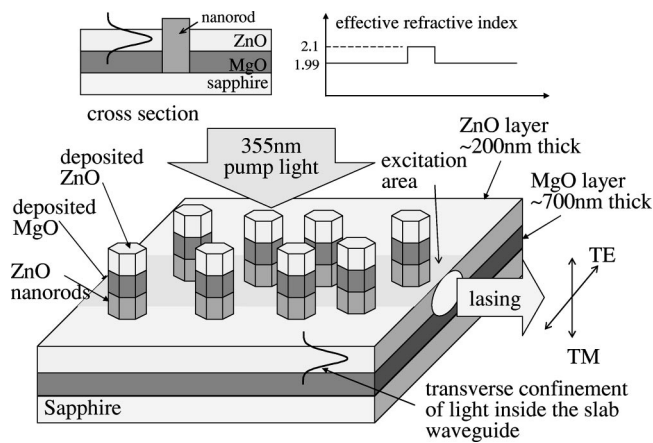[a)]Author to whom correspondence should be addressed; electronic mail: esfyu@ntu.edu.sg

FIG. 1. Schematic diagram of the ZnO nanorods embedded in the ZnO epilayers. Bottom half of the diagram shows the arrangement of optical excitation of the sample. Top half of the diagram shows the cross section of the sample and the corresponding effective refractive index of the slab waveguide and ZnO nanorods.

be obtained simultaneously so that random laser action with coherent feedback can be achieved.

Room-temperature optical characteristics of ZnO nanorods embedded in ZnO thin film were studied under optical excitation. Optical pump was achieved by using a cylindrical lens to focus a pump stripe of length 5 mm and width 60 μm on to the sample. A polarizer in the direction perpendicular [transverse electric (TE)] and parallel [transverse magnetic (TM)] to the thin-film surface was also used to analyze the polarization properties of the lasing light. Figure 2 shows the light–light curves and emission spectra of the sample. Threshold pump intensities of TE and TM polarizations are found to be about 800 kW/cm^2 and 1.6 MW/cm^2, respectively. For both polarizations, single-broad emission spectra with a FWHM of about 15 nm are observed. When the excitations exceed the threshold, sharp lasing peaks of a linewidth less than 0.4 nm are emerged from the single-broad emission spectra. A further increase in pump intensity increases the number of lasing modes as the increase in optical gain excites more cavity modes with higher losses. The generation of single-broad emission spectra above threshold is due to the weak optical feedback from the facets of the slab waveguide.[7] This indicates that the FP modes will not be supported in the slab waveguide otherwise the single-broad emission spectra will be suppressed. The excitation of shape lasing peaks is in fact caused by the formation of closed-loop paths for light through recurrent scattering (i.e., coherent feedback). It can be shown that the average separation between nanorods is less than 90 nm and the field reflectivity between the nanorods and slab waveguide is about 0.03. Hence, the estimated scattering mean-free path will be less than the emission wavelength so that the claim of recurrent scattering is justified. In addition, the lasing intensity of TM polarization is weaker than that of TE polarization, which is due to the orientation of the ZnO nanorods (i.e., the scattering strength is strong for TE polarization). The high threshold pump intensities of our sample compared with that of the ZnO nanorod arrays embedded in anodic Al$_2$O$_3$ (Ref. 4) may be due to the low optical reflectivity (i.e., scattering strength) at the boundaries between the ZnO nanorods and the slab waveguide. However, a direct comparison between the two
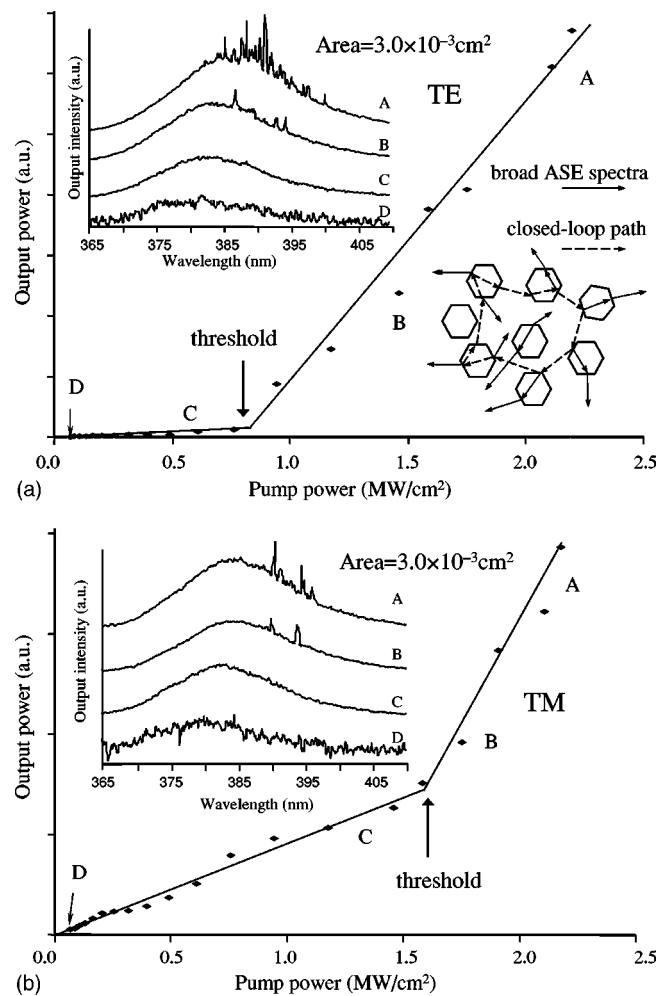

FIG. 2. Light–light curves and emission spectra of (a) TE polarization and (b) TM polarization form the ZnO nanorods embedded in ZnO epilayers. The insets on the top-left-hand side corner is the emission spectra at various pump intensities and that on the bottom-right-hand side corner is a schematic diagram showing the formation of closed-loop path for light through recurrent scattering (dashed arrow) and single-broad ASE spectra (solid arrow) in the sample.

cases may not appropriate, as the threshold pump intensity is also dependent on the excitation volume.

Figure 3 plots the emission spectra measured at different observation angles. It is noted that different emission spectra can be observed in different directions. This is because different laser cavities formed by multiple scattering can have different output directions so that lasing spectra observed at different angles are different. Furthermore, the dependence of random laser action on the excitation area was studied. Figure 4 shows the variation of emission spectra with excitation area (from 5×10^{-4} cm^2 to 3×10^{-3} cm^2) for constant pump intensity of 2.6 MW/cm^2. It is observed that before the critical area (i.e., 1×10^{-3} cm^2) is reached, no sharp lasing peak is observed. However, when the excitation area has exceeded the threshold, sharp lasing peaks with a linewidth of less than 0.4 nm appear. The number of sharp lasing peaks increases with the increase of excitation area. This is because in a large excitation area, more closed-loop paths for light can be formed. As a result, random laser action could occur in more cavities formed by recurrent scattering. However, if the excitation area was reduced to below a critical size, laser oscillation stopped. This is because if the
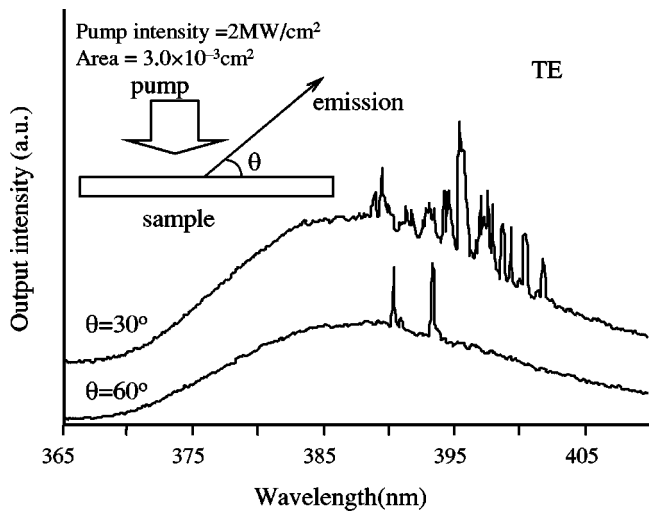
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FIG. 3. Spectra of TE polarization radiate from 30° and 60° from the sample surface. The pump intensity is 2 MW/cm^2 and the excitation area is kept at 3×10^{-3} cm^2.

closed-loop paths are too short, the amplification along the loops is not high enough to achieve lasing.

From the above measurement, all of the essential characteristics of random laser action have been observed from the sample. Furthermore, the random laser theory has shown 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 $V \sim (\ell_s \ell_g)^{3/2}$.[3] It is noted that ℓ_g is defined as twice the inverse of the optical gain, g, of the ZnO epilayers. In Ref. 7, it has been shown that g of ZnO thin films is roughly proportional to the pump intensity so that the empirical expression of ℓ_g can be written as $\ell_g^{-1} \sim 1/2(\partial g/\partial P) P_{th}$, where P_{th} is the threshold pump intensity and $\partial g/\partial P$ is a constant. Hence, it is possible to write $A_{th}^{2/3} \sim 2\ell_s d^{-2/3}(\partial g/\partial P)^{-1} P_{th}^{-1}$. This linear relationship between $A_{th}^{2/3}$ and P_{th}^{-1} indicated that the lasing threshold characteristics of the ZnO nanorods embedded in ZnO epilayers agreed well with the random laser theory.[3] Figure 5 plots the varia-
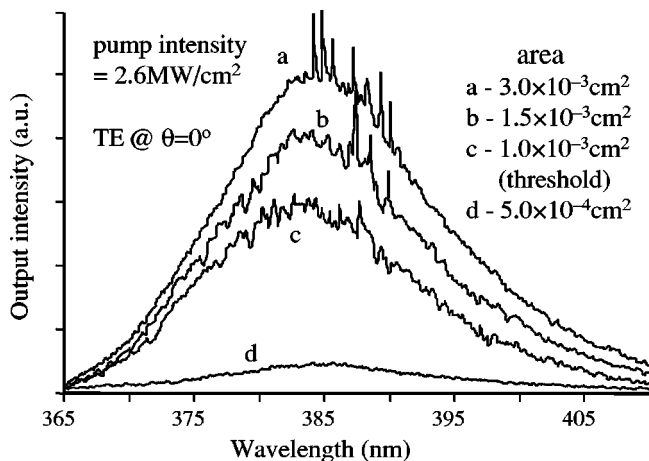

FIG. 4. Spectra of TE polarization of the sample when the excitation area is (from top to bottom) 3.0×10^{-3}, 1.5×10^{-3}, 1.0×10^{-3}, and 5.0×10^{-4} cm^2. The excitation intensity is 2.6 MW/cm^2.
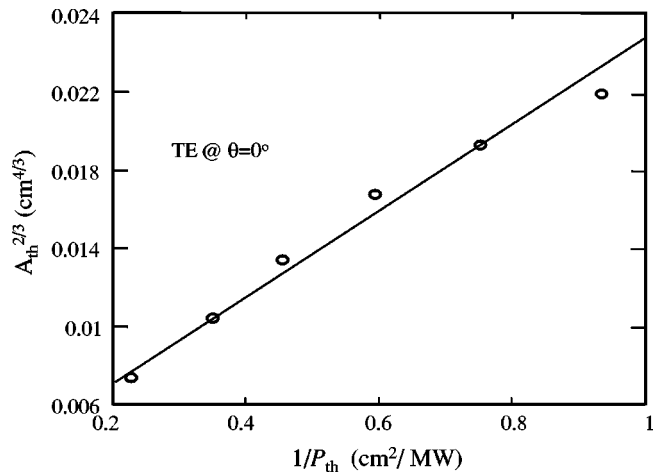

FIG. 5. Plot of $A_{th}^{2/3}$ versus P_{th}^{-1} of the sample under TE polarization emission.

tion of $A_{th}^{2/3}$ against P_{th}^{-1} for the TE polarization. It is observed that $A_{th}^{2/3}$ versus P_{th}^{-1} exhibits a linear relationship so that the lasing mechanism of the ZnO nanorod arrays embedded in the ZnO epilayers is verified to be random laser action.

In conclusion, the FP cavities formed between the top and bottom natural facets of our ZnO nanorods will not sustain lasing caused by the short cavity length (i.e., very high cavity loss). Furthermore, the random laser action is also not detected due to the low density of the ZnO nanorod arrays. However, if the ZnO nanorod arrays are embedded in the ZnO epilayers, which consist of a thick MgO buffer layer and a ZnO thin film, random lasing can be obtained. This is because the presence of ZnO epilayers increases (reduces) the optical gain (the scattering mean-free path) so that random lasing can be sustained. In addition, the dependence of threshold pump intensity on the excitation area agrees well with the random laser theory. Hence, the claim of random lasing action can be supported in ZnO nanorod arrays is verified.

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