Suppression of Random Lasing Modes in Polycrystalline ZnO Thin-Film by Using Distributed Bragg Reflector

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***Abstract*—Formation of vertical-cavity random laser by using a polycrystalline ZnO thin-film optically coupled to 11.5 pairs of Al_2O_3–ZnO multilayer distributed Bragg reflector (DBR) is proposed. Lasing characteristics of the laser observed from the surface of the polycrystalline ZnO thin film are studied without and with the presence of DBR. It is found that the corresponding spectral linewidth can be limited to about 4 nm at high excitation intensity under the influence of DBR.**

***Index Terms*—Distributed Bragg reflectors (DBRs), random laser, zinc oxide.**

I. Introduction

ULTRAVIOLET (UV) random laser action has been observed in mirrorless polycrystalline ZnO thin films under optical excitation at room temperature [1], [2]. As the fabrication of UV lasers using polycrystalline ZnO thin films can avoid the difficulty in realizing cleaved facets to sustain optical feedback, the choice of polycrystalline ZnO thin films may facilitate the development of practicable and low-cost UV semiconductor lasers. However, high scattering loss and a large number of random modes limited the usefulness of ZnO thin-film random lasers for practical applications [3]. Although the use of ridge waveguides was proposed to reduce the scattering loss of ZnO thin-film random lasers, many random modes remained especially at a high excitation power [4]. On the other hand, the fabrication of photonic crystal structure in ZnO thin films may be the only possible way to realize narrow linewidth UV lasing at the expense of stringent fabrication requirements [5].

In this letter, we propose the realization of vertical-cavity random lasers by depositing a polycrystalline ZnO thin film on to a distributed Bragg reflector (DBR). The polycrystalline ZnO thin film, which supports random lasing action at $\sim$385 nm, forms an active layer of the vertical-cavity random laser. The thickness of the polycrystalline ZnO thin film and the reflection spectrum of the DBR are designed in such a way that the peak lasing wavelength of the polycrystalline ZnO film falls into the first bandedge energy on the high energy side of the reflection spectrum of the vertical-cavity random laser. As a result, random modes that radiate away from the first bandedge energy (i.e., within the bandgap region of the highest reflectivity) will be suppressed. Hence, the number of random modes being excited will be restricted to a small value. In fact, the design of vertical-cavity random lasers is similar to that using 2-D photonic crystal to control the lasing wavelength of microcavity lasers [6]. This is because the DBR structure is a photonic crystal in 1-D.

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II. Fabrication Procedures

Fig. 1(a) shows the schematic diagram of a vertical-cavity random laser. The DBR consists of 11.5 pairs of $\lambda/4$ Al_2O_3 and ZnO dielectric layers, which were deposited on a quartz substrate by a modified filtered cathodic vacuum arc (FCVA) system. The FCVA system has two sources which are capable of depositing two types of metal–oxide films sequentially. Zn and Al metal targets, both with purity of 99.99%, were used to form Zn and Al plasmas, respectively, from the two independent cathodic arc sources [7]. Oxygen gas is introduced from an outlet which is 2 cm above the surface of the substrate. The gas is mixed with the metal plasmas to form metal–oxide thin film. The thickness of the metal–oxide thin films can be controlled by a gold crystal thickness monitor. In our studies, oxygen flow rate was set to 160 sccm and oxygen partial pressure was maintained at 5×10^{-4} torr. The arc currents for the Zn and Al cathodic vacuum arc sources were kept at 60 and 160 A, respectively. The substrate was set to room temperature with a rotational speed of 30 rev/min. The deposition rate of the Al_2O_3 and ZnO thin films were found to be 1.0 and 1.3 nm/min, respectively.

The DBR was designed and fabricated with peak reflectivity of 99% at the wavelength of 405 nm. It is noted that the required thickness for Al_2O_3 and ZnO dielectric layers are 59.5 and 48.5 nm, respectively. Fig. 1(b) shows the transmission electron microscopy (TEM) image of the DBR realized by the FCVA system at room temperature. It is observed that the interfaces between Al_2O_3 and ZnO are relatively smooth. The measured and calculated reflection spectra of the DBR are plotted in Fig. 2. It is observed that both the measured and calculated results are in good agreement. Hence, this indicated that the FCVA technique is suitable to fabricate optical-quality metal–oxide thin films even at room temperature.

There are three reasons to fabricate the DBR using ZnO and Al_2O_3 as the dielectric materials: 1) The contrast of refractive

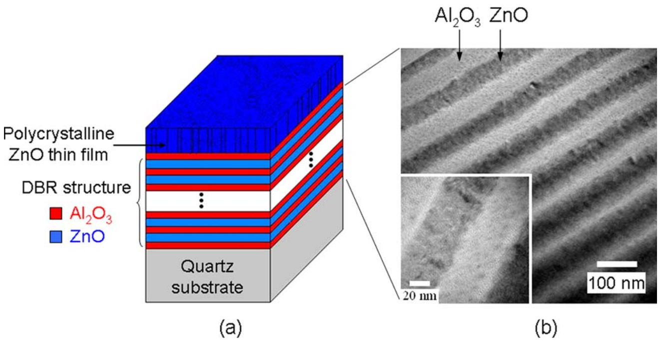

Fig. 1. (a) Schematic of the vertical-cavity random laser. (b) TEM image of the Al_2O_3–ZnO DBR fabricated by the FCVA technique.

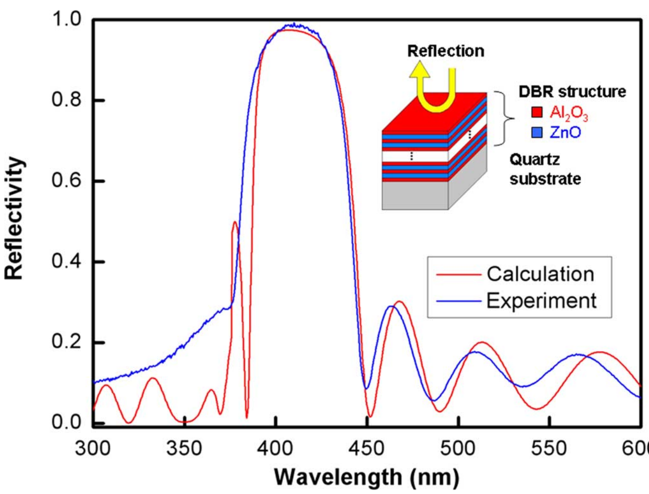

Fig. 2. Normal reflection spectrum of the DBR—comparison between measurement and calculation.

index between ZnO ($n = 2.1$) and Al_2O_3 ($n = 1.77$) is relatively large (i.e., 0.33 at 385 nm) when compared to the other materials that can be grown by the FCVA technique. Therefore, the number of dielectric pairs required to achieve 99% of reflectivity can be less than 12 pairs. 2) Al_2O_3 has the lowest absorption at around 385 nm among other available metal–oxide thin films. Furthermore, although the lasing emission wavelength of the polycrystalline ZnO thin films is near 385 nm, ZnO dielectric layers of the DBR only exhibit strong bandedge absorption at or below 380 nm [2]. Hence, the use of Al_2O_3–ZnO DBR to confine random modes inside the polycrystalline ZnO thin films is realistic. 3) By limiting the use of target materials to Al and Zn, the corresponding deposition process via FCVA technique can be simplified. Hence, mass production of UV ZnO random lasers at a low-cost is possible.

It must be noted that the as-grown ZnO thin-film fabricated by the FCVA technique at room temperature does not support random lasing action [7]. Hence, the polycrystalline ZnO thin film, which supported random lasing action, has to be fabricated with deposition conditions different from that of the DBR. In the fabrication, the polycrystalline ZnO thin film, which has a thickness of 300 nm, was deposited on the DBR with substrate temperature set to 300 °C. This thickness of the polycrystalline ZnO film will ensure the first bandedge energy matches with the peak gain wavelength of the polycrystalline ZnO thin film.

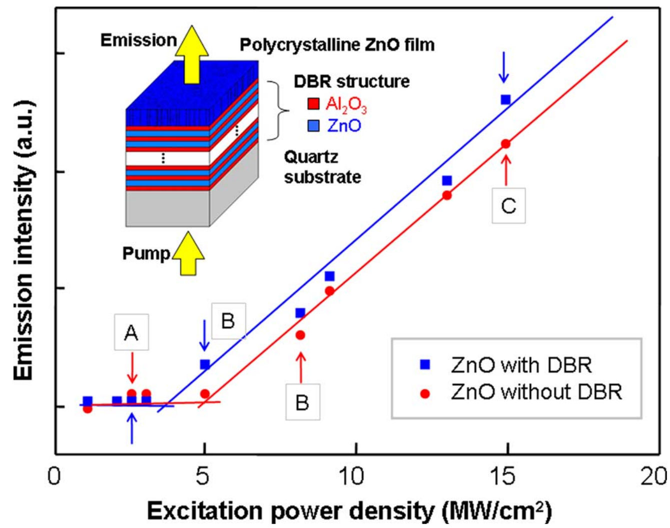

Fig. 3. Light–light curves of the polycrystalline ZnO thin films with and without DBR structure. The samples were excited normal to the surface of the quartz substrates and the emission light was measured perpendicular from the surface of the polycrystalline ZnO films.

During the deposition, the oxygen flow rate and partial oxygen pressure were maintained at 177 sccm and 7×10^{-4} torr, respectively. An arc current of 80 A was used to produce the polycrystalline ZnO thin films with better optical-quality than that of the DBR. It can be shown that the polycrystalline ZnO thin film deposited at 300 °C can sustain random lasing action at wavelength of about 385 nm. On the other hand, although the DBR experienced post-growth annealing at 300 °C during the deposition, its reflection spectrum remained unchanged after the deposition [4]. In addition, random lasing action was not supported inside the DBR structure. Another polycrystalline ZnO thin film with thickness of 300 nm was also realized on quartz substrate by the FCVA technique for comparison.

III. Experimental Results

The room-temperature optical characteristics of the polycrystalline ZnO films with and without DBR were studied under optical excitation by a frequency-tripled Nd : YAG (yttrium aluminum garnet) laser (355 nm) at pulsed operation (120 ps, 10 Hz). Optical pumping was achieved by using a spherical lens to focus a 1-mm diameter pump spot on the quartz substrate in order to simplify the experimental setup. Hence, light emitted from the surface of the polycrystalline ZnO films were analyzed. Fig. 3 shows the light–light curves of the polycrystalline ZnO films with and without DBR. It is observed that a kink occurs at about 5.1 MW/cm^2 (3.3 MW/cm^2) for the sample without (with) DBR. The absorption of excitation power at the quartz substrate and DBR structure had been subtracted from the light–light curves. It is noted that the presence of DBR reduces the lasing threshold of the random lasers by about 1.5 times.

Fig. 4(a) shows the emission spectra measured from the surface of the film without DBR at room temperature. For pump intensities exceeding 5.1 MW/cm^2, sharp peaks (i.e., random modes) are excited at around 385 nm. The mechanism of radiative recombination inside the polycrystalline ZnO thin film may

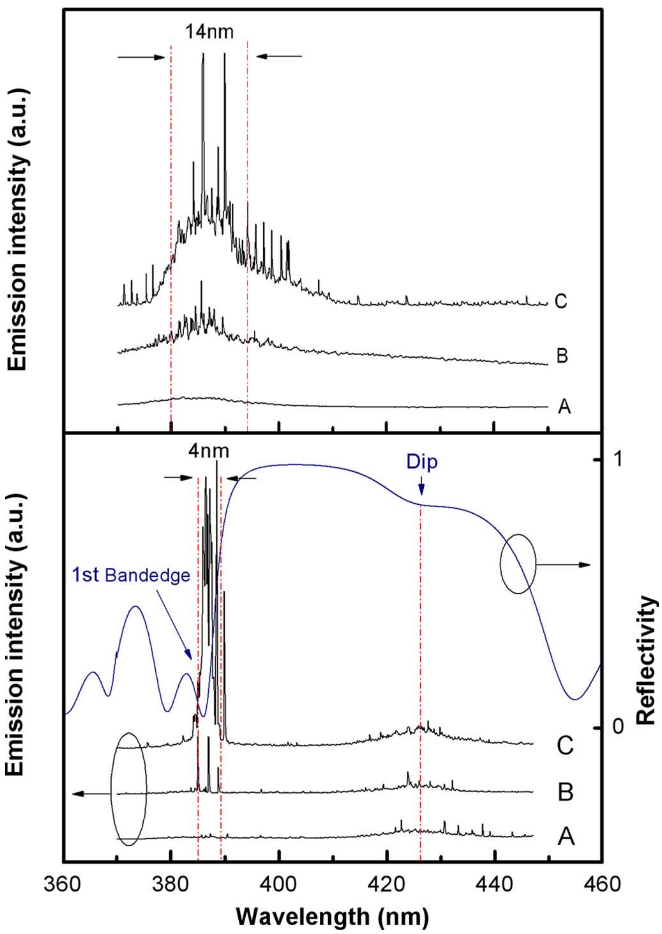

Fig. 4. Emission spectra measured perpendicular from the surface of the polycrystalline ZnO films (a) without and (b) with DBR structure. The normal reflection spectrum measured from the polycrystalline ZnO film/DBR is also shown in (b).

be attributed to the process of exciton–exciton scattering [2]. The excitation of narrow sharp peaks and the present of the kink in the light–light curves suggested that the lasing process is due to coherent random lasing action as there is no other feedback mechanism can sustain optical feedback [4]. For pump intensities at about 2.9 times its threshold, full-width at half-maximum (FWHM) of the lasing spectrum increases from 4 to more than 14 nm. This indicates that the number of random modes increase with the increase of pump intensities. This is in fact one of the lasing characteristics of random lasers [1].

Fig. 4(b) shows the emission spectra of the film with DBR. The reflection spectrum of the polycrystalline ZnO thin film on the DBR is also plotted in the figure. It is observed that the first bandedge energy of the reflection spectrum is located at 385 nm. A small dip also appears at a wavelength of around 425 nm. This is because the presence of polycrystalline ZnO thin film modified the reflection spectrum of the DBR. Strong random lasing action is only observed near the wavelength of the first bandedge energy. This is because the peak gain wavelength of the polycrystalline ZnO film matches with the first bandedge energy of the vertical-cavity random laser. Furthermore, the FWHM of the lasing spectrum is limited to 4 nm even at 4.5 times of its threshold. However, the region of the highest reflectivity, which is the bandgap of the vertical-cavity random laser, strongly suppressed the excitation of lasing emission. On the other hand, a weak spontaneous emission is observed at the wavelength of about 425 nm (i.e., at the dip of the reflection spectrum). The excitation of spontaneous emission is due to the defects of ZnO in the DBR after post-growth annealing at 300 °C.

The reduction of threshold and narrowing of lasing spectrum can be explained by the resonance of random modes within the vertical cavity which formed between the air–polycrystalline ZnO interface and the DBR. Hence, the DBR provides a high reflection to recycle the scattered light back into the polycrystalline ZnO film so that the corresponding threshold can be reduced. Furthermore, the vertical-cavity modifies the reflection spectrum of the DBR. As a result, the spectral width of the lasing emission can be limited by the bandwidth of the first bandedge spectrum.

IV. CONCLUSION

A simple one-step fabrication process by using the FCVA technique was proposed to fabricate a polycrystalline ZnO thin film on a Al_2O_3–ZnO multilayer DBR. The polycrystalline ZnO on DBR, in which the polycrystalline ZnO film acts as the active layer, formed a vertical-cavity random laser. The lasing behavior from the surface of the polycrystalline ZnO without and with DBR was studied at room temperature. It is shown that the random laser with DBR exhibit UV random lasing at around 385 nm with spectral width limited to 4 nm even at 4.5 times its threshold. In addition, the corresponding threshold of the laser can be reduced by 1.5 times.

REFERENCES

[1] H. Cao, Y. G. Zhao, X. Liu, E. W. Seelig, and R. P. H. Chang, "Effect of external feedback on lasing in random media," *Appl. Phys. Lett.*, vol. 75, pp. 1213–1215, 1999.
[2] Y. Chen, N. T. Tuan, Y. Segawa, H.-J. Ko, S.-K. Hong, and T. Yao, "Stimulated emission and optical gain in ZnO epilayers grown by plasma-assisted molecular-beam epitaxy with buffers," *Appl. Phys. Lett.*, vol. 78, pp. 1469–1471, 2001.
[3] X. Jiang and C. M. Soukoulis, "Time-dependent theory for random lasers," *Phys. Rev. Lett.*, vol. 85, pp. 70–73, 2000.
[4] C. Yuen, S. F. Yu, S. P. Lau, and C. K. Chen, "Design and fabrication of ZnO light-emitting devices by using filtered cathodic vacuum arc technique," *J. Cryst. Growth*, vol. 287, no. 1, pp. 204–212, 2006.
[5] X. Wu, A. Yamilov, X. Liu, S. Li, V. P. Dravid, R. P. H. Chang, and H. Cao, "Ultraviolet photonic crystal laser," *Appl. Phys. Lett.*, vol. 85, pp. 3657–3659, 2004.
[6] M. M. Muller, A. Bauer, T. Lehnhardt, and A. Forchel, "Widely tunable photonic crystal coupled cavity lasers on GaSb," *IEEE Photon. Technol. Lett.*, vol. 20, no. 13, pp. 1100–1102, Jul. 1, 2008.
[7] C. Yuen, S. F. Yu, E. S. P. Leong, S. P. Lau, K. Pita, H. Y. Yang, and T. P. Chen, "Room temperature deposition of p-type Arsenic doped ZnO polycrystalline films by laser-assist filtered cathodic vacuum arc technique," *J. Appl. Phys.*, vol. 101, pp. 094905 1–7, 2007.