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Optically and electrically pumped random lasing from ZnO films annealed at different temperatures

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ABSTRACT

We have investigated the optically and electrically pumped random lasing (RL) actions in ZnO films annealed at low and high temperatures respectively. While the optically pumped RL threshold for the ZnO film annealed at a low temperature, which features stronger light scattering and larger optical loss, is far higher than that for the ZnO film annealed at a high temperature, the electrically pumped RL threshold currents for the two ZnO films are almost the same with the electrical pumping scheme of metal–insulator–semiconductor structure. It is suggested that if the lasing region within the ZnO film is narrow enough in the case of electrical pumping, the strong light scattering can substantially alleviate the adverse effect of large optical loss on the onset of RL.

1. Introduction

Random lasing (RL) from ZnO has been expended extensive research efforts since the first report of optically pumped RL from ZnO powders by Cao et al. [1,2]. In the past decade, RL actions in ZnO materials of different forms have been realized by optical pumping [3–5], which is, however, not desirable for practical applications. Yu et al. opened the way to enabling RL from ZnO materials by means of electrical pumping. They reported electrically pump RL from the stripes of ZnO clusters embedded in SiO_2 film sandwiched in *p*-GaN (SiC) and *n*-ZnO:Al [6]. Subsequently, our group achieved the electrically pumped RL from ZnO polycrystalline film which acted as the semiconductor component in a metal–insulator–semiconductor (MIS) structure [7]. Of late years, a diversity of strategies has been successively proposed to realize the electrically pumped RL from the ZnO films [8–10]. As for the correlation between the optically and electrically pumped RL actions in the ZnO films, it has been rarely addressed. Intuitively, if the optical pumping threshold for a ZnO film is high, one tends to believe that the electrically pumped RL from this ZnO film is consequently difficult to achieve. Whether such an intuition is true under all circumstances needs to be essentially clarified.

In this work, we have investigated the optically and electrically pumped RL actions from two ZnO films annealed at different temperatures. It is found that the ZnO film annealed at a high temperature can be optically pumped into RL with a far lower threshold than that annealed at a low temperature. While, based on the pumping scheme using the MIS structure, the two ZnO films can be electrically pumped into RL at nearly the same threshold current. The mechanism underlying the above-mentioned results has been tentatively elucidated.

2. Experimental

The ZnO films with a thickness of ~150 nm were deposited on heavily arsenic-doped silicon (n^+–Si) substrates by reactive DC sputtering. They were subsequently annealed at 750° C for 1 h and 1100° C for 1 min under O_2 ambient, denoted as ZnO films A and B respectively for the sake of description. Each annealed sample was cut into two halves. One was used for the optically pumped RL, the other was used to act as the semiconductor component in a MIS structured device for the electrically pumped RL. The preparation procedures for the ZnO films and MIS structured devices can be referred to our previous report [11]. In the case of optical pumping, the samples placed in thin quartz test tubes were pumped by third harmonics of the two-stage Q-switched Nd:YAG laser (355 nm, ~6 ns, 15 Hz) with a pumping area of ~3 mm^2. The pumping energy density was varied from 9 to 200 mJ/cm^2. The photoluminescence (PL) spectra were acquired by a photo multiplier (PET-79). In the case of electrical pumping, the MIS structured devices were forward biased with the negative voltage connecting to the n^+–Si substrate.

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The wavelength scanning size was 1 Å for acquiring the electroluminescence (EL) spectra.

3. Results and discussion

Fig. 1(a) and (b) show the typical scanning electron microscopy (SEM) images for the ZnO films A and B, respectively. As can be seen, the crystal grains of ZnO film B are much larger. It is understandable that growth of crystal grains in the ZnO film is facilitated by the annealing at higher temperature. Fig. 1(c) illustrates the PL spectra of ZnO films A and B. Comparatively, the ZnO film B exhibits much stronger emission in both ultraviolet (UV) and visible regions, indicating that this ZnO film has a much smaller amount of non-radiative recombination centers. Apparently, as shown in Fig. 1(a) and (b), the ZnO film B has much fewer grain boundaries, where non-radiative recombination usually takes place. Moreover, it is believed that the point-like defects that contribute to non-radiative recombination could also be fewer in the ZnO film B than in the ZnO film A [12]. In a word, it can be believed that the optical loss is less in the ZnO film B. On the other hand, multiple light-scattering proceeding within the ZnO film A is much stronger. Evidently, there are far more grain boundaries in the ZnO film A than in the ZnO film B. Thus, the light is much more frequently subjected to scattering by the grain boundaries in the ZnO film A.

Fig. 2(a) and (b) show the room temperature (RT) PL spectra in the UV region for the ZnO films A and B, respectively, pumped by different laser intensities. As for the ZnO film A, as shown in Fig. 2(a), with increasing pumping intensity and therefore ever-higher density of excitons in ZnO, the emission band becomes more broad and red-shifted, which is characteristics of electron–hole plasma emission [13]. At the pumping intensity of 200 mJ/cm^2, there is still no obvious lasing peak appeared in the emission spectrum. Regarding the ZnO film B, even at quite a low pumping intensity of 9 mJ/cm^2, the emission band becomes narrower than that of the ZnO film A pumped with the intensity of 18 mJ/cm^2. With the pumping intensity increased to 18 mJ/cm^2, the emission band is further narrowed and a number of sharp peaks are observed. The sharp peaks in the spectra can be ascribed to RL from the ZnO film B. Therefore, it can be concluded from Fig. 2 that the optically pumped RL threshold for the ZnO film B is much lower than that for the ZnO film A. This is due to that the optical loss is less in the ZnO film B, as mentioned above.

Fig. 3(a) and (b) show the RT EL spectra acquired at different injection currents for the two kinds of MIS structured devices (herein the ZnO films A and B are used as the light-emitting layers, respectively). The two devices are named after devices A and B hereafter. As can be seen, all the spectra feature a number of discrete sharp peaks with a linewidth less than 5 Å and with uneven spacing between two neighboring peaks, indicating the occurrence of RL. It is worthy to mention that there are much more sharp peaks, i.e. lasing modes in the spectra of the device A than in those of the device B. This indicates that there are more 'random cavities' formed in the device A, which results from the stronger multiple light scattering proceeding in the ZnO film A.

Fig. 4 shows the dependences of the detected output optical power on the injection current for the devices A and B, respectively. In our measurement configuration, only ~2% of the output power from the devices was detected. For either device, a threshold behavior being characteristic of lasing action is exhibited. That is, the output power increases much rapidly with the injection current beyond a threshold. It can be seen that that the electrically pumped RL threshold currents for the two devices are almost the same. With increasing injection current, the detected output power from the device B is larger than that from the device A.

So far, it has been demonstrated that while the optically pumped RL threshold for the ZnO film A is far larger than that for the ZnO film B, the two ZnO films can be electrically pumped into RL by the pumping scheme of MIS structure with almost the same threshold current. Such results above will be explained in the following.

In the case of optically pumped RL, the excitation light can penetrate into the ZnO films. Thus, the emitted light is subjected to multiple scattering while propagating throughout the films. Considering the ZnO films in this work are quite thin, the effect of scattering on the depth profile of the excitation light intensity across the ZnO film can be neglected. Accordingly, with the same pumping intensity, the densities of excitons in the ZnO films A and B are nearly identical. This leads to nearly equivalent optical

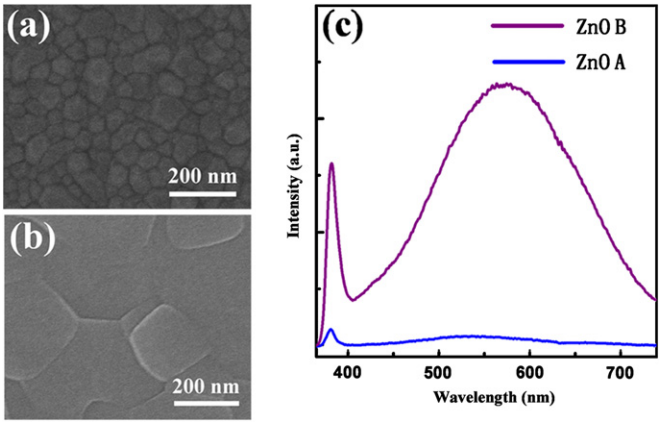

Fig. 1. (a) and (b) Typical plane SEM images of ZnO films A (annealed at 750 °C for 1 h) and B (annealed at 1100 °C for 1 min), respectively. (c) PL spectra of ZnO films A and B.

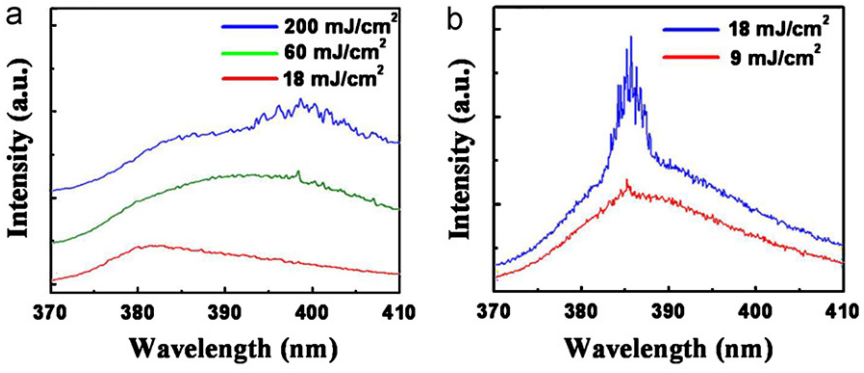

Fig. 2. (a) and (b) PL spectra acquired with different pump intensities for the ZnO films A (annealed at 750 °C for 1 h) and B (annealed at 1100 °C for 1 min), respectively.

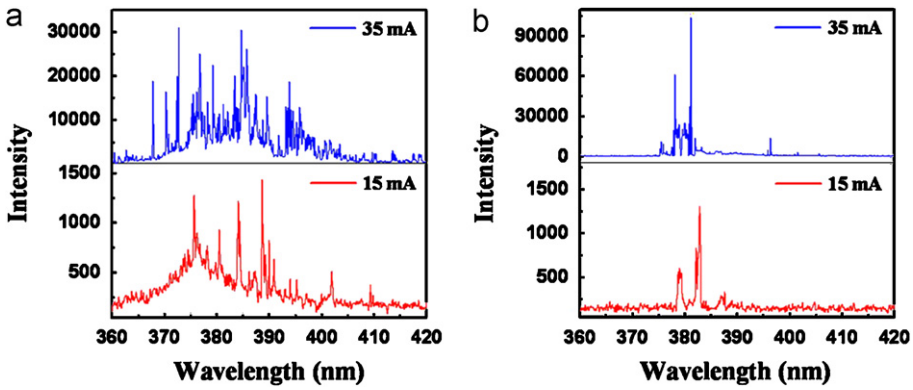

Fig. 3. (a) and (b) EL spectra for the MIS structured devices A and B using the ZnO films annealed at 750 °C for 1 h and 1100 °C for 1 min as the light-emitting layers, respectively, under different injection currents.

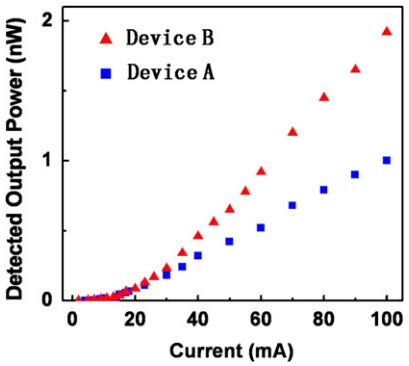

Fig. 4. Dependences of detected output power on the injection current for the devices A and B, respectively.

gain factors in the two ZnO films. However, as mentioned above, the ZnO film A is of larger optical loss. If the light experiences scattering paths of equivalent length in the two ZnO films, it is evident that the optical gain picked up in the ZnO film A is relatively smaller. Therefore, a larger intensity is needed to optically pump the ZnO film A into RL.

In the case of electrical pumping by means of MIS structure, as pointed out in our previous reports [14,15], substantial injected carriers accumulate in the region nearby the SiO_2/ZnO interface, where stimulated emission (SE) and therefore optical gain will be achieved above a threshold current. This satisfies a fundamental condition for RL [16]. The emitted light is concurrently subjected to multiple scattering in the SE region. RL will occur if the achieved optical gain is larger than the optical loss after multiple scattering events [17]. Returning to the comparison of the two ZnO films A and B, which act as the light-emitting layers in the two MIS structured devices, we can see the following scenarios. On one hand, as mentioned above, the multiple light scattering in the ZnO film A is stronger than that in the ZnO film B. Our recent work has defined the compensation between optical gain and light scattering in the electrically pumped RL by means of MIS structure [15]. Therefore, if only considering the effect of light scattering, the ZnO film A can be electrically pumped into RL by means of MIS structure at a lower threshold current. On the other hand, the optical loss in the ZnO film A is larger than that in the ZnO film B. This leads to a far larger threshold intensity for the optically pumped RL from ZnO film A, as described above. While, in the case of electrically pumped RL achieved in the MIS structure, the SE region is very narrow, with a limited number of non-radiative recombination centers therein. Besides, the results obtained in Ref. [18] also indicate that the scattering loss in ZnO film can be reduced by the coating layer on it. Therefore, the optical loss does not exert disadvantageous effect in the electrically pumped RL as strong as that in the case of optically pumped RL. In a word, taking into account of concurrent light scattering and optical loss, we can derive that the RL threshold currents for the ZnO films A and B could be comparable in the case of electrical pumping scheme using the MIS structure. Actually, this derivation has been verified by the experimental result as shown in Fig. 4. With increasing injected current, the SE region within the ZnO film becomes progressively wider, thus involving more and more non-radiative centers. As a result, the detrimental effect of optical loss on the RL is not ignorable any more as the ZnO-based MIS structured devices are fed with sufficiently large currents. As revealed in Fig. 4, with increasing injection current above a critical value, the detected output power of RL from the MIS structured device A, which is of larger optical loss, becomes increasingly smaller than that from the counterpart device B.

4. Conclusions

In summary, we have investigated the optically and electrically pumped RL actions in the ZnO films annealed at low and high temperatures, respectively. By comparison, the ZnO film annealed at the low temperature possesses stronger light scattering and larger optical loss. The optically pumped RL threshold for the ZnO film annealed at the low temperature is much higher than that for the counterpart annealed at the high temperature. Nevertheless, the electrically pumped RL threshold currents for the two ZnO films are almost the same in the pumping scheme of MIS structure. This work hints that if the lasing region within ZnO film in the case of electrical pumping is sufficiently narrow, the strong light scattering can offset the disadvantage effect of large optical loss on the onset of RL.

Acknowledgments

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