The formation characteristics of closed-loop random cavities inside highly disordered ZnO polycrystalline thin films

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(Received 28 December 2005; accepted 28 February 2006; published online 24 March 2006)

The formation characteristics of closed-loop random cavities inside highly disordered ZnO films with and without rib waveguide structure are investigated. The size dependence of the random cavities inside the random media on temperature and pump intensity profile is studied by applying Fourier transform to the corresponding lasing spectra. Simple rate equation analysis has revealed that the formation of the random cavities depends mainly on the profile of the optical gain, which is a function of the pump intensity profile and carrier diffusion length of the random media. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2189908]

Coherent random lasing has been observed in highly disordered ZnO polycrystalline thin films due to the formation of closed-loop random cavities.[1] The selection of high-quality (i.e., low loss and high gain) scattering paths, which controls the size and location of the random cavities, has been suggested to be the formation mechanism.[2] Hence, it is concluded that the available size of the random cavities is determined by a critical cavity length[3] and it is strongly related to the material properties of the random media. However, this conclusion is based on the assumptions that (1) the pump excitation is uniformly distributed over the random media and (2) the small diffusion length of carriers has an insignificant influence on the profile of optical gain inside the random media, as the excitation area is usually relatively large.[4,5] In this letter, we show that the excitation pump, whose intensity distribution has a Gaussian profile, modifies the optical gain of the random media so that the size and location of the random cavities are also affected. In addition, the optical gain profile of the random media with a rib waveguide structure can be altered by carriers with small diffusion length as the nonradiative recombination centers increase at the sides of the rib waveguide. Therefore, the formation characteristics of the closed-loop random cavities are not determined mainly by the material properties of the random media.

The highly disordered ZnO films, with ZnO ($\sim$180 nm)/SiO_2 ($\sim$400 nm)/Si structure, were fabricated by the filtered cathodic vacuum arc technique and followed by thermal annealing at 900 °C for 2 h. Rib waveguides with a fix length of $\sim$0.1 cm and different widths (2.5 and 6.5 μm) were defined on the ZnO films by ion-beam etching. Detailed fabrication procedures of the rib waveguide random lasers have been described elsewhere.[6] The lasing characteristics of the ZnO films was studied under optical excitation by a frequency-tripled Nd yttrium aluminum garnet (Nd:YAG) laser (at 355 nm) at pulsed operation (6 ns, 10 Hz). The optical pump was focused using a cylindrical lens to a stripe of length $\sim$1 mm and width $\sim$40 μm on the surface of the ZnO films. The lasing emission was measured from the edge of the films. It must be noted that the intensity profile of the pump stripe has a Gaussian profile with beamwidth of $\sim$18 μm.

Figure 1 shows the lasing spectra of the ZnO films with (of width 2.5 μm) and without rib waveguide structure. Uniform excitation is obtained from the ZnO film with rib waveguide structure. It is observed that the lasing spectra redshift with the increase in temperature T due to band-gap shrinkage.[7] In addition, the number of lasing peaks decreases with the increase in T especially for the film with rib waveguide structure. This implies that the number of random cavities formed inside the random media is reduced with the increase in T. As the pump intensity remains unchanged, the

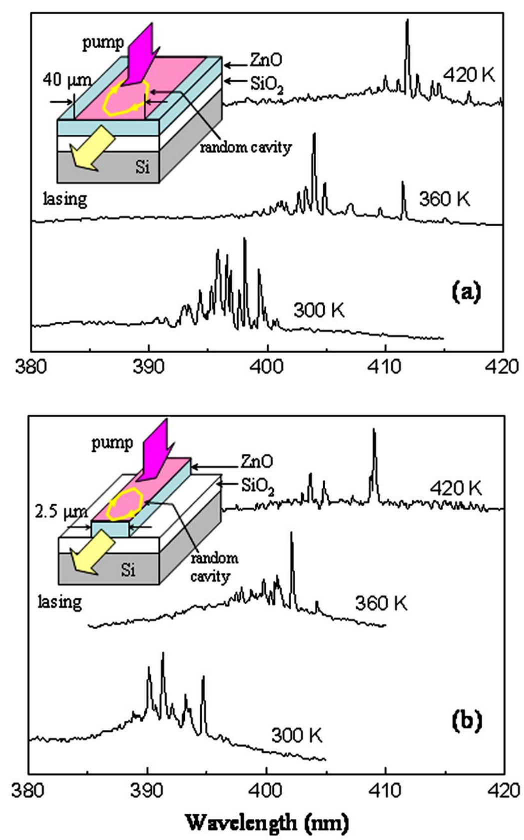

FIG. 1. (Color online) Lasing spectra for the highly disordered ZnO films (a) without and (b) with a rib waveguide structure at pump intensities of $\sim$0.4 and $\sim$0.6 MW/cm^2, respectively, for different temperatures.

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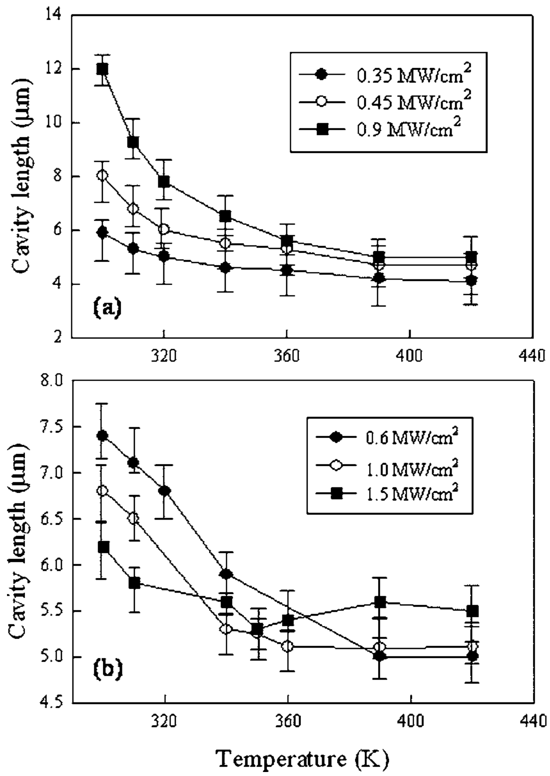

FIG. 2. Fundamental cavity length (obtained from Fourier transform to the lasing spectra) versus temperature under different pump intensities for highly disordered ZnO films (a) without and (b) with a rib waveguide structure.

optical gain of the random media reduces with the increase of T. This is because the corresponding threshold pump intensities increase exponentially with T (i.e., the characteristics temperature of both ZnO films are found to be ~95 K). Therefore, lasing peaks due to random cavities with relative higher cavity loss will first disappear from the emission spectra. However, it is uncertain whether the random cavities, which are assumed to have circular shape, with large or small random cavities will be suppressed at high T. However, we believe that the formation of the closed-loop random cavities depends mainly on the optical gain profile, which is a function of T and pump intensity, inside the random media. Hence, the influence of T and pump intensity on the size of the random cavities is investigated to further understand the corresponding formation mechanism.

We performed Fourier transform of the lasing spectra to evaluate the dominant cavity length of the random cavities.[3] Figure 2 plots the fundamental cavity length L versus T at different pump intensities for the ZnO films with and without rib waveguide structure. The threshold pump intensities of the random media with and without rib waveguide structure are equal to ~0.33 and ~0.26 MW/cm^2, respectively, at T =300 K. It is observed that L reduces with the increase in T for the case without a rib waveguide. In addition, the increase in pump intensities increases the value of L. However, for the case with rib waveguide [i.e., Fig. 2(b)], the dependence of L on T and pump intensities is more complicated. In general, L reduces with the increase in T. However, L reduces (increases) with the increase in pump intensities for T lower (larger) than 340 K. We have repeated the experiment for a rib waveguide laser with a width of 6.5 μm, and found that the dependence of L on T and pump intensities is similar to that of ZnO films without rib waveguide structure. From the experiments, it is noted that (1) the size of the random cavities depends strongly on the magnitude and profile of the optical gain inside the random media and (2) the presence of waveguide structure will alter the formation of random cavities only if the width of the waveguide is equal or shorter than 2.5 μm.

In order to further understand the formation mechanism of the closed-loop random cavities, we studied the distribution of carrier concentration (i.e., equivalent to studying the optical gain profile) over the pump region of the random media. This can be done by solving the one-dimensional (1D) rate equation of carrier concentration N, given as $\partial N/\partial t = I - N/\tau_n + D_n \partial^2 N/\partial x^2$, where x is the distance across the pump stripe, I is pump intensity, τ_n (=1 ns) is the carrier lifetime, D_n $(=L_n^2/\tau_n)$ is the diffusion coefficient, and L_n is the diffusion length. It is noted that the pump beam has a Gaussian profile (all the commercial available frequency-tripled Nd:YAG lasers have output beam of a Gaussian profile, however, most of the lasing experiments reported in the literatures have ignored this fact)[8] so that the pump intensity can be written as $I = I_o \exp(-x^2/\omega^2)$, where I_o is the peak pump intensity and ω (=9 μm) is the radius of the Gaussian beamwidth. For the case of ZnO films without rib waveguide structure, it is assumed that N decreases exponentially outside the pump region with the increase of x. On the other hand, for the ZnO films with a rib waveguide structure, N at the side of the waveguide has to satisfy the boundary condition $\partial N/\partial x + \nu_{sur} N/D_n = 0$, where ν_{sur} $(=1\times 10^3$ cm/s) is the surface recombination velocity. It is noted that the 1D rate equation of N is accurate enough to study the threshold characteristics of the random cavities as the influence of stimulated recombination is not significant.

Figure 3(a) plots the normalized values of $N(x)$ versus x for the random media without a rib waveguide and only half of the distribution of N is shown in the figures as they are symmetric at $x=0$. We assumed that the value of L_n used in the calculation ranging from 0.1 to 1.0 μm. This is because the presence of ZnO grains inside the random media confines the diffusion of carriers to a distance of 0.1 μm (i.e., minimum size of grains).[9] On the other hand, it is reported that the value of L_n in ZnO films can be from ~0.5 to ~1.0 μm for T increasing from 300 to >600 K, respectively.[10] It is observed that for a reduction of L_n from 1.0 to 0.1 μm, there is an increase in N at $x<6.5$ μm, especially at the middle $(x=0)$ of the excitation region. The optical gain of the random cavities (G) can be approximated linearly by $G \approx A^{-1}\int a_N[N(x)-N_o]|\Psi(x,y)|^2 dxdy$, where A is the modal area, a_N is gain coefficient, N_o is the carriers at transparency and $\Psi(x,y)$ is the optical field profile of the random cavities. The expression of G indicates that the optical gain (1) is high at region near to the middle of the pump stripe and (2) can be improved for the random media with small value of L_n.

Figure 3(b) plots the normalized values of N versus x for the films with rib waveguide structure of width equal to 2.5 μm. It is observed that the distribution of optical gain across the rib waveguide is less dependent on the intensity distribution of the pump beam. However, the presence of nonradiative recombination centers at the side of the rib waveguide affects the uniformity of the optical gain especially for the case with small L_n. In fact, this is the reason the

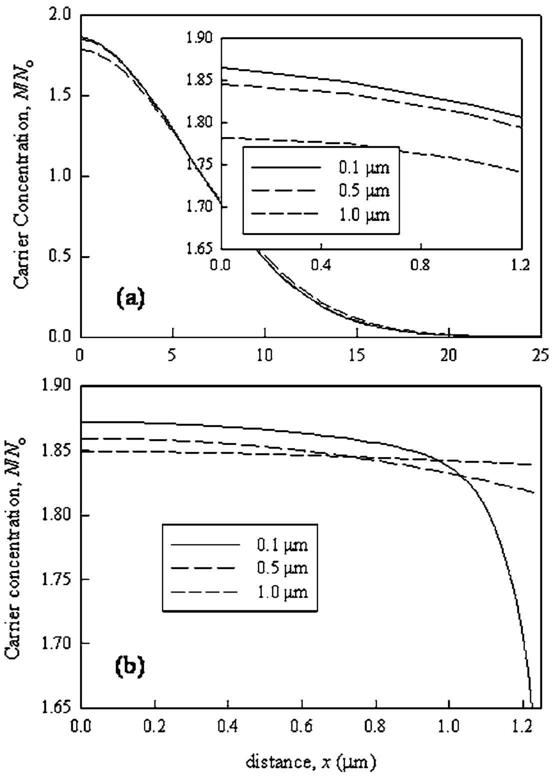

FIG. 3. Normalized carrier concentration versus distance across the pump stripe for different values of L_n for highly disordered ZnO films (a) without and (b) with a rib waveguide structure.

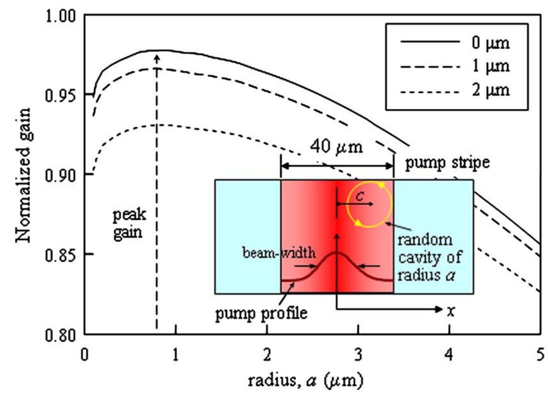

FIG. 4. (Color online) Normalized average gain versus the radius of the circular closed-loop random cavities at distance c away from the middle of the pump stripe for the highly disordered ZnO films without a rib waveguide structure.

formation mechanism of closed-loop random cavities in ZnO films with narrow rib waveguide is different to that without rib waveguide (i.e., as indicated in Fig. 2). For L_n=0.1 μm (equivalent to the rib waveguide tested at $T<320$ K), the optical gain is significantly reduced (when compared to that without rib waveguide) near the side of the rib waveguide, especially at high pump intensities. Therefore, L is restricted to a small value due to the large difference in optical gain between the regions near the middle and the side of the rib waveguide. One the other hand, the optical gain is more uniform for the large L_n (equivalent to the rib waveguide operating at $T>360$ K). In this case, the increase of L with pump intensity as observed from Fig. 2(b) at $T>360$ K is due to the small difference in gain between the regions near the middle and the side of the rib waveguide.

The expression of G for the random media without a rib waveguide structure can be evaluated by assuming that the random cavities have a closed-loop circular shape with radius a at a distance c from the middle of the pump stripe and $N(x) \approx N_b \exp(-x^2/\omega^2)$, where N_b is a constant. Hence, G can be expressed as $G \approx (2\pi a \delta)^{-1}[g(a+\delta, c) - g(a, c)]$, where $g(a,c) = 2a_N \int_{c-a}^{c+a} \sqrt{a^2-(x-c)^2}[N_b \exp(-x^2/\omega^2) - N_o]dx$ and δ is the equivalent width of $|\Psi|^2$. Figure 4 plots the normalized average gain $G/(a_N N_b)$ of the random cavities versus a for different c. It is observed that the normalized gain has a maximum at $a = \sim 0.8$ μm, which is equivalent to $L \sim 5$ μm. This value is close to that the measured value of L for the random media at high T [see Fig. 2(a)]. This shows that at high T where G is reduced, only random cavities of size ~ 5 μm (i.e., coinciding with the peak gain) will have sufficient gain to sustain lasing. Hence, this also explains the reason the reduction of L ceases at a constant value for $T > 360$ K. This is because the increase of T reduces G so that only the size of closed-loop random cavities match the peak gain (shown in Fig. 4) will still have sufficient gain to sustain lasing. It is also observed from Fig. 4 that the value of G reduces with the increase of c (i.e., equivalent to the formation of random cavities away from the middle of the pump stripe). This indicates that the closed-loop random cavities should form near to the middle of the pump stripe, as the corresponding average optical gain is higher than the other region.

In conclusion, we have found that the Gaussian profile of pump intensity, which is usually ignored in the study of lasing action in ZnO films, has significant influence on the size and location of the closed-loop random cavities inside the random media. The formation of random cavities can also be modified by a small value of L_n, whose existence is often neglected in the consideration, for the random media with a small rib waveguide structure. Furthermore, the formation of the size and location of the closed-loop random cavities is shown to be dependent on the profile of the optical gain inside the random media. This implies that the lasing characteristics of the random media can be controlled. Hence, our understanding of the formation characteristics of random cavities will help to design ZnO random lasers to achieve better operation efficiency to suit practical applications.

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