Design and fabrication of zinc oxide multilayers light emitting devices

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DESIGN AND FABRICATION OF ZINC OXIDE MULTILAYERS LIGHT EMITTING DEVICES

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SUMMARY

Zinc Oxide (ZnO) exhibits a wide band-gap of $\sim 3.4\text{eV}$ and a large exciton binding energy of $\sim 60\text{meV}$. ZnO has shown a large potential in the development of Ultraviolet (UV) light emitting devices, operating at room temperature. However, the crystal structure of ZnO has hindered its advancement in UV conventional lasers due to the difficulty in facet-cleaving, results in large scattering loss and high laser threshold.

An alternative solution is in ZnO Random Laser. It is a laser that relies on multiple scatterings rather than conventional facets to create feedback, and therefore, the performance of the laser will not be obstructed by its crystal structure. Still, ZnO Random Lasers suffer from several disadvantages that limit its full capability, such as high laser threshold, lack of directionality and multimode operation.

In this thesis, the improvement of ZnO radiation emission efficiency by integrating with multilayers structure is discussed, particularly with ZnO Random Laser. To fabricate ZnO-based multilayer device in this project, a double-arm Filtered Cathodic Vacuum Arc (FCVA) system is built. Various optimizations have performed to produce high quality ZnO thin films, with processes such as fine-tuning of the arc current and altering the guiding magnetic fields. To achieve high accuracy in thickness control, various thickness-monitoring methods were installed and tested. Eventually, success is

made when metal oxide is deposited with an accuracy of 1nm in thickness.

For ZnO emission characterization, a low temperature photoluminescence setup is built. The cooling is achieved with a close circuit cyro-chamber where liquid nitrogen is used as our heat exchange agent. This enables us to measure photoluminescence response at as low as 77k. We have used ND:YAG laser at 355nm as our excitation source and a monochromator with photomultiplier tube for our signal capturing.

A theoretical model based on the transfer matrix method is developed for the design of Distributed Bragg Reflectors (DBR). We have chosen ZnO:Mg and Al_2O_3 as our reflective materials for our DBR design. Meanwhile, the model is modified and applied on the studies of another complex multilayer structure, the anti-resonant reflecting optical waveguide (ARROW) vertical-cavity surface-emitting laser. Various losses under high current injection were investigated and a new proposed structure with modified refractive index profile is suggested as a possible solution.

Lastly, experimental implementation of multilayer structures on ZnO is performed. A Vertical Cavity, Surface Emitting Random Laser is designed and fabricated. This particular design has successfully narrowed the random laser emission spectrum up to 2.5 times even under high excitation intensity. Furthermore, the complete structure is fabricated with one-step FCVA technique that provides a possible low cost solution for commercial UV lasers.

ZnO Multiple Quantum Wells (MQW) device is also fabricated with our FCVA system under low substrate temperature condition ($<300^{\circ}\text{C}$), yet, its performance is still comparable with most ZnO MQW currently fabricated by other methods. This will open up a new possibility of coating ZnO MQW lasers on various different structures, disregarding the lattice matching condition and the consideration of its melting point.

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Chapter 1

INTRODUCTION

1.1 Backgrounds and Motivation

Since 1970s, semiconductor laser has found its important role in the communication industry. The first optical communication system was put into service at 1976, operating over 11 km of fiber at 45Mbit s^{-1} [1]. Previously, voice communications were mainly operated with wired phone and television through broadcasting. Without the introduction of the optical communication system, it is almost impossible to have high-definition television, video-on-demand access, broadband Internet and transmit terabits of information in the split of a second over transcontinental distance in today's world.

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Unique properties of semiconductor lasers, such as high power output, small spot size and coherence emission in nature have made them ideal for optical communication. Also, unlike other form of lasers, semiconductor lasers can be designed with well-controlled and narrow-linewidth optical emission that can minimize the effect of dispersion in optical fiber with an extremely high modulation rate ^[2].

Besides optical communications, semiconductor lasers have also found their applications in optical data storage, displays, and bio-sensing because of its compactness in size, small power consumption, long lifespan, large volume production and low manufacturing cost ^[3].

Furthermore, short wavelength semiconductor lasers are predicted to be essential in the next generation high-compact optical data storage systems. Today, many researchers have foreseen the potential of Ultraviolet (UV) semiconductor lasers in the field of communications, defense, water purification and chemical processing ^[4].

1.1.1 Choice of UV Material

In order to obtain UV emission, large bandgap material ($>3\text{eV}$) is required, such as diamond^[5], SiC^[6], BN^[7], GaN^[8], ZnO^[9] and ZnS^[10]. Yet, only ZnO, GaN and ZnS are direct-bandgap materials, which are favorable for radiation recombination. This is essential for light emitting devices when efficiency is one of the key considerations.

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Among the above direct-bandgap materials mentioned, GaN is the most popular used in the manufacturing of commercial laser diode, mainly because of the successful realization in p-type GaN, which is critical for electrical excited light emitting devices ^[11,12]. However, most of the fabrication methods are toxic and require high energy for its deposition process, such as high substrate temperature ($\sim 1000^{\circ}\text{C}$) or the assistance of electron cyclotron resonance ^[13]. Furthermore, GaN is inert to most of the chemical that make post-deposition processes such as the fabrication of the waveguide, a more difficult task. Hence, manufacturing cost for GaN-based optoelectronic devices is still relatively high and this has hindered the penetration of GaN in the commercial markets.

Conversely, although ZnO is still suffered from the difficulty of p-type doping, it may become another low cost substitute for UV optoelectronics industry in the near future if this technical challenge can be overcome. Similar with GaN, ZnO has a large bandgap of $\sim 3.4\text{eV}$, which is capable for UV emission. Also, since ZnO is an oxide-based semiconductor, high quality ZnO ^[14,15,16,17] can be deposited at a temperature lower than most of the nitride based semiconductor material ^[8]. Furthermore, ZnO has a large exciton binding energy of 60meV , a binding energy that is almost 2 times larger than GaN ($\sim 28\text{meV}$) and 2.4 times of the thermal energy in room temperature. This suggested that the realization of high power room temperature operated excitonic UV laser could be possible. Recently, a research group from Japan has successfully reproduced p-type ZnO ^[18], which may overcome the challenge of fabricating ZnO electronic devices.

Table 1-1 listed a comparison of some intrinsic properties between GaN and ZnO.

	GaN	ZnO
Structure	Wurtzite	Wurtzite
Bandgap (eV)	3.4 (R.T.)	3.37 (R.T.)
Exciton Binding Energy (meV)	28	60
Lattice constant (Å)	a=3.186 c=5.178	a=3.249 c=5.206
Refractive index (n)	~2.4-2.5	~2.1
Thermal expansion coefficient (°K)	a=5.59×10-6 c=7.75×10-6	a=6.51×10-6 c=3.02×10-6

Table 1-1 Comparison between GaN and ZnO

1.1.2 ZnO lasers

Even though optically pumped ZnO laser has been realized earlier by cleaving facets on ZnO with sapphire substrate to form cavities^[19], its Wurtzite crystal structure has resulted in a large scattering loss and therefore, the laser threshold is high. Hence, this is not economic for commercial production^[20].

On the other hand, random lasers^[21] can provide an alternative solution for the manufacturing of low cost ZnO lasers. Random laser does not require conventional resonators to provide feedback and therefore, it does not require a high-quality facet to sustain lasing. Instead, random lasers depend on multiple scatterings as their feedback, which is created by the scatterers within its medium.

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Although random laser has its advantage of being a mirrorless laser, it has its drawback, too. The emission of the random laser is not directional ^[22] and its cavities created by multiple scatterings imposed high optical loss ^[23]. The laser threshold is found to be higher and the laser emission has multimodes. Although various researches have provided solutions to the high threshold and non-directional emission by fabricating different types of waveguide structures ^[24,25] and nanostructures ^[26,27], the problem with wide spectrum, multimodes emission is still unavoidable. As such, it is necessary to look into ways to reduce the multimodes operation of random lasers and yet, achieving low pumping threshold and high gain lasers.

1.2 Objectives

The objectives for this thesis are listed as below:

- To investigate the technique of fabricating complex multilayer structure for the application in UV light emitting devices.
- To perform a theoretical study of the effect of the multilayer structure on the performance of lasers.
- To improve the lasing efficiency with the multilayer structure (i.e., reduce threshold, improve the directionality and coherence of emission light) of random lasers.

1.3 Major Contributions

This research work is focused on the study of ZnO random lasers based on disordered films, which were fabricated with the Filtered Cathodic Vacuum Arc (FCVA) technique. Optical characterization has been employed for the investigation of the optical properties of the samples. The main contributions of the work done for this thesis are as follows:

For the deposition of the multilayer metal-oxide structure, a double-arms FCVA system is built. Various modification and optimizations with the deposition system have performed to produce high-quality ZnO thin films. In order to deposit metal oxide with high accuracy in thickness, several methods for thickness-control are installed and tested. Also, simulation and experimental

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testing have performed to reduce the majority of noises, for a better control in deposition thickness.

For the characterization of the optical properties of ZnO thin film, a low temperature photoluminescence setup is also assembled. Various troubleshooting have performed on the laser and also the design of the heat exchange circuit.

Theoretical model based on transfer matrix method is developed to study the optical behavior of Bragg reflectors. Meanwhile, the model is modified and applied on the studies of the Antiresonant Reflecting Optical Waveguide (ARROW) Vertical-Cavity Surface-Emitting Laser (VCSEL). Various losses have examined and a possible solution is proposed.

To improve the performance of ZnO random lasers, Distributed Bragg Reflector (DBR) has proposed to be incorporated with the ZnO polycrystalline thin film. The reflection spectrum of the DBR is designed to match the gain spectrum of the ZnO. Both of the DBR structure and ZnO gain medium are fabricated with a single-step FCVA method, which provides a possible low cost solution for narrow emission random lasers.

Lastly, an experimental realization of Multiple Quantum Wells (MQW) structure for low threshold ZnO emission is achieved. The device is fabricated under a low substrate temperature condition with FCVA and its radiation

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characteristics are studied. This accomplishment has created a new possibility to integrate ZnO low threshold lasers with various low melting point and lattice-mismatched substrates.

1.4 Organization

This thesis is organized as follows: In chapter 2, a general background and some of the optical properties that are associated with the objectives of this thesis are first discussed. A brief introduction of ZnO laser is given and follows by a section on ZnO random lasers. Detailed analysis of recent research particularly in the field of ZnO random laser is then provided. Basic fundamental principles and properties are also mentioned together with some of its possible applications. Chapter 3 presents the fabrication method of the metal oxide. We have chosen FCVA technique as our fabrication method and its working principles is discussed. Various troubleshooting and modification to our FCVA system are documented as well. Also, the discussion of the thickness controlling method is included in the later part of this chapter. In chapter 4, the setup of our low-temperature photoluminescence characterization system is presented. It is followed by a low temperature characterization study of the stimulated emission from the FCVA deposited ZnO polycrystalline thin films. Next, a theoretical model, developed for the design of DBR is covered in chapter 5. This model is also applied on a study with ARROW structure later. In chapter 6, the design and the realization of ZnO Vertical Cavity Surface Emitting Random Laser is covered. The characterization is performed with our photoluminescence system and results are compared with ZnO thin film. In Chapter 7, experimental realization of ZnO multiple quantum wells structure fabricated with FCVA is achieved, also, the experimental results are compared and discussed. The work presented in the thesis is summarized in chapter 8 and

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recommended future works are discussed.

Chapter 2

LITERATURE REVIEW

2.1 Zinc Oxide

ZnO has gained substantial interest in semiconductor research because of its wide band gap ($\sim 3.4\text{eV}$) and also its large exciton binding energy ($\sim 60\text{meV}$). These made ZnO a favorable candidate for UV semiconductor laser, especially for room-temperature operation. As such, we have chosen ZnO as our platform for random lasers studies in this thesis.

Researches on ZnO can be traced back to many decades. Material characterizations such as lattice parameters were investigated as early as 1935 [28,29,30,31,32,33]. Also, optical properties and processes in ZnO were studied intensively. [34,35,36,37,38,39] One of the main reasons that attracted attentions to ZnO is because of its large exciton binding energy, which paves the way for an intense near band edge excitonic emission at room or even higher temperature. This is because the binding energy is almost 2.4 times larger than the thermal

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energy at room temperature. Besides, ZnO is very resistive to high-energy radiation ^[40,41,42,43] and amenability to wet chemical etching that allows easy fabrication for micro devices ^[44]. Furthermore, the crystal structure of ZnO is very similar with GaN, in which, technologies for GaN can be easily implemented ^[45].

ZnO is a II-VI compound semiconductor with its ionicity falls in between the covalent and ionic semiconductor. Mainly, it has 3 types of crystal structure; Wurtzite, Zinc Blende and Rocksalt. In ambient environment, the thermodynamically stable phase is Wurtzite, which possesses those desirable properties for UV lasers. Zinc Blende structure can only be found when ZnO is grown on cubic structure substrate and Rocksalt can only be obtained at high pressure. Furthermore, the transformation from Wurtzite structure to Rocksalt structure is reversible and the transformation pressure is found to be ~9 GPa ^[46]. Therefore, most of the ZnO thin films in our studies are having a Wurtzite structure.

The conduction band of the Wurtzite-structured ZnO is s-like with Γ_7^c symmetry whereas its valence band is p-like state and it is split into 3 bands caused by the influence of the crystal field and spin-orbit interactions ^[47]. Hence, the near band edge absorption and emission are mainly dominated by the transitions between these bands. Usually, the transition between the conduction band and the 3 valance bands are denoted by A, B and C, which are also referred as heavy hole, light hole and crystal-field split orbit respectively.

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Teke *et al* ^[48] has performed some detail investigations with the intrinsic optical transition in ZnO by low temperature photoluminescence. He was able to observe the A-free excitons and their first excited-state transition. Together with the energy separation of ground-state, excited-state peak position and the assumption of hydrogen-like exciton, the A-free exciton binding energy was predicted to be 60meV with a corresponding band gap energy of 3.4 eV at 10 K.

Besides the free exciton transition, bound excitons transition also has a strong role in ZnO optical transition mechanism. Bound excitons transitions are extrinsic and they are associated with dopants and defects, which create addition discrete electronic states within the bandgap. Depends on the purity of ZnO, donor- bound-exciton (DBE) and acceptor-bound-exciton (ABE) are normally observed between 3.348eV to 3.374eV ^[49].

Another type of optical transition reported in ZnO is known as two-electron satellites (TES). This transition involves a radiative recombination between an exciton and a neutral donor, which causes the donor to be at its excited state. This will result in a transition energy that is lesser than the DBE energy by the different of the first excited state and the ground state of the donor. This transition is reported in the spectral region between 3.32 to 3.34eV ^[50,51].

In addition to those exciton-related transitions mentioned, observation of emission peak at about 3.2eV together with other few emission peaks were also

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reported ^[50]. These emission peaks were concluded to be the transitions of donor-acceptor-pair (DAP) and Longitudinal-optical (LO)-phonon replicas respectively.

2.2 ZnO Laser

2.2.1 Conventional laser ^[52]

Laser is well known for its spatially, temporally and spectrally coherent radiation emission. It is also directional with intensity stronger than any other conventional sources of light, such as fluorescent lamp. To achieve lasing, two requirements are essential, the gain condition and the phase condition.

Firstly, the gain in the optical medium must be greater than its loss; otherwise absorption will disallow light amplification to occur. Hence, population inversion is needed and this can be achieved by an external pumping either through optical excitation or electrical excitation.

Secondly, its phase is also necessary to be matched (i.e., phase $\sim m \times 2\pi$ where m is an integer), such that stimulated emission can be dominant over spontaneous emission. Phase matching can be achieved by creating a cavity to provide optical feedback. Usually, the gain medium of the conventional laser is enclosed within a resonant cavity, which is made of reflective elements. Smooth surface is necessary such that scattering loss can be minimum, which is detrimental to the lasing operation. Generally, reflectivity surface of the semiconductor laser cavity can be formed via cleaved, etched facets, deposited reflective metal films at the edges or with an external pair of mirrors. To enhance the light confinement in semiconductor laser, optical waveguide design is commonly deployed.

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There are mainly 3 types of resonance modes in a conventional laser structure, i.e. the longitudinal mode, the transverse electric mode (TE) and the transverse magnetic (TM) mode.

The longitudinal mode is determined by the length of the laser cavity and it is associated with the integrals multiples of half wavelength. i.e., $m = \lambda/2nL$, where m is the mode number, L is the length of the cavity, λ is the optical wavelength and n is the refractive index of the medium. Depends on the finesse of the cavity, the existence of these modes is commonly represented by narrow and sharp peaks with full width at half maximum of < 1 nm in the emission spectrum.

While the longitudinal mode is depended on the length of the cavity, TE and TM modes rely on the cross sectional area of the laser structure. The TE (TM) mode is parallel (perpendicular) to the active layer while propagation of light (k) is along the active region. Conventionally, the TE mode will lase first because of its stronger structural optical confinement that results in a lower gain threshold.

In ZnO, the stimulated emission at low excitation intensity is mainly induced by exciton-exciton scattering process. The emission energy usually occurs at about 3.15eV ^[53], which is below the free exciton energy by the sum of the exciton binding energy and the mean kinetic energy $\frac{3}{2}K_B T$ ^[54,55]. This emission is the

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result of the inelastic collision between excitons, causing an exciton to be excited to a higher state with a photon of the energy difference between the free exciton, the binding energy of the exciton and the thermal energy.

When the excitation intensity is high, the density of exciton increases, their wave functions begin to overlap and the exciton starts to lose its individual character due to phase space filling and coulomb interaction. The stimulated emission mechanism is then transformed from exciton-exciton scattering into electron-hole plasma (EHP) process. The density, which EHP occurs is known as the “Mott density” and it is given by

$$n_m = \frac{K_B T}{2a_B^3 E_B^{ex}}$$

where a_B is the exciton Bohr radius and E_B^{ex} is the exciton binding energy. Mott density for ZnO is found to be about $3 \times 10^{23} \text{ m}^{-3}$ [56].

Generally, EHP emission for ZnO occurs at about 3.1 eV. The emission spectrum become broaden and red shifted as the excitation intensity increases due to band gap renormalization [57].

As we have mentioned earlier, ZnO crystal structure is not favorable for facet-cleaving. Although reports [58,59] have shown the successful realization of ZnO conventional laser, the laser threshold is too high for any commercial lasers application and it is not economically incline with the current market. Therefore, an alternate laser mechanism is discussed in the following section.

2.2.2 ZnO Random Lasers

Conventional semiconductor lasers have been studied extensively and were well established. Various methods to improve their performance, such as enhancing their optical confinement with different types of waveguide structures have been proposed and realized. More complex and sophisticated structures such as the Distributed Bragg Reflector (DBR), Distributed Feedback (DFB), quantum well and Vertical-Cavity Surface-Emitting Lasers (VCSELs) for single mode operation have also been studied theoretically and experimentally broadly.

However, as the laser application advanced towards the UV region, conventional optical feedback method becomes less effective. Metallic mirror has poor reflectivity at frequency beyond its plasma frequency and scattering losses increase as a function of wavelength reduction. Furthermore, the easiness of cleaving smooth facets is dependent on its material structure. For the case of ZnO, the Wurtzite structure created challenges in obtaining smooth facets surface via cleaving.

Recently, a non-conventional type of laser, random laser is evolved. It is a mirrorless laser that does not depend on any conventional optical feedback. It relies of its disordered medium to create multiple scatterings for coherent feedback and as well as optical amplification. As such, random laser provides an alternative and yet, an easier and cheaper method for UV lasers fabrication.

2.2.2.1 Development of Random Lasers

In 1966, Ambartsumyan *et al* first proposed a different type of laser with a non-resonant feedback by replacing one of the mirrors in the laser cavity with a highly scattering medium ^[60], in which, it demonstrated a very strong interaction between the lasing modes^[61]. Later in 1967, Letokhov *et al* ^[62] predicted theoretically that multiple light scattering in a negative absorption medium is possible to produce lasing, especially when the scattering mean free path is significantly shorter than the sample size. Random Lasers was realized experimentally in 1970, when Varsanyi *et al* observed stimulated emission in PrCl_3 and PrBr_3 particles ^[63].

First ZnO related random laser was reported in 1981, when stimulated emission is detected from ZnO powder ^[64]. Also, Lawandy *et al* ^[65] observed a spectral narrowing in the emission spectrum and the emission behaviors exhibit laser like threshold dependence characteristics, which is known as Amplified Spontaneous Emission (ASE) later, in random laser theory. Most of the earlier detailed study of UV stimulated emission in ZnO is primarily due to Cao *et al* and he described the ASE as incoherent random lasing when their group discovered coherent random laser action in ZnO powder^[66]. This has inspired many interests in investigating the role of particles in gain medium ^[67,68]. Presently, various experiments and modeling of the random laser theory have been devised to investigate the properties of random lasers ^[69,70,71,72].

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In the early works, laser dye solutions with particles as scatterers were used to study the fundamental properties of random lasers ^[33,73]. However, this is not practical because the particles sediment and sometimes agglomerate in the solution over time. Therefore, solid-state medium has proposed, which is based on nano-composite films of either laser with scatterers ^[74,37,75] or active scatterers in passive medium ^[76].

Most of the time, random lasers suffer high lasing threshold, multimode and non-directional emission. Therefore, various researches have performed to improve the efficiency of random lasers.

One of the more common methods will be to incorporate waveguide design with random laser. This will improve its optical confinement and result in a better directional emission laser ^[77,78]. For ZnO, MgO is proposed for the fabrication of ZnO ridge waveguide laser and result shows a great improvement in the directionality of the transverse beam profile ^[75].

Another possibility will be to apply external feedback, such as placing a mirror closely to the sample edge ^[79,80] or coating the facet with metal to improve its reflectivity ^[75]. Experimental results demonstrated that if the mirror is not too far away from the sample edge, the lasing threshold can be reduced as much as half ^[76].

2.2.2.2 Working Principles of Random Lasers

Analogous with conventional laser, random laser requires the fulfillment of the gain condition and the phase condition. Like conventional lasers, population inversion in the medium is needed and therefore, positive gain is necessary. However, instead of having reflective facets to produce coherent feedback, random laser uses multiple scattering in a disordered medium to achieve its phase matching requirement, as shown in figure 2-1. When the scattering is sufficiently strong, close loop scattering paths may form and hence, providing coherent optical feedback.

Figure 2-1. Schematic diagram of a disordered material. (a) Formation of closed-loop path for light through recurrent scattering in the amplifying and scattering particles. (b) Photon triggering the generation of another photon (amplification) before leaving the medium.

The degree of scattering depends on the size of the scatterers and also the contrast of the refractive index between the scatterer with the medium.

Basically, scattering consists of 3 regimes; the Rayleigh, Mie and Geometric optics, ^[81] depends on the particle size. The size parameter, x , is given as $x = 2\pi r/\lambda$, where r being the radius of the particle. In general, when $x < 0.1$, it is consider as the Rayleigh regime, where $0.1 < x < 50$ is the Mie regime and $x > 50$ is the geometric regime.

Usually, strong scattering only occurs in Mie regime where the size of the scatterer has about the same magnitude as the wavelength. In Geometric regime, the scattering cross section, σ_{sc} , a parameter that quantify the degree by which a particle scatters light, is too big for its geometric area and in the Rayleigh regime, the scatterer is too small to be effective. Generally, σ_{sc} also increases with the refractive index contrast between the scatterer and the medium.

2.2.2.3 Coherent vs Incoherent Random Lasers

As mentioned earlier, there are 2 types of random lasers; incoherent and coherent feedback. Incoherent random laser does not have a close loop scattering path, i.e. Light does not return to its starting point after a round trip and therefore, its emission is not as define as a typical laser. Yet, narrowing in the emission spectrum can still be observed, i.e., ASE, because multiple scatterings lengthen the traveling path of the photon and trigger stimulated emission. The light amplification is given by $I = I_0 e^{gl}$, where g is the optical gain in the medium and l is the path length traveled by the photon in the

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medium^[66, 82, 83]. Although such laser does not have a distinctive lasing emission spectrum, a “kink” in the light-light characteristic curve shows the non-linear behavior similar to that of the laser threshold.

When there is a sufficient degree of randomness in the medium, it is possible for a photon to scatter within the medium and return to its starting point^[84]. When it happens, various closed loop scattering paths are formed and created multiple random cavities within the gain medium. With enough gain at the close loop path, lasing occurs and its lasing modes are at integral multiples of half-wavelengths of each random cavity. Lasing peaks emerge and increase with the increase of the excitation intensity. The presence of multiple defined lasing peaks and a “kink” in the light-light characteristic curve is the distinct signatures for coherent random lasers.

Properties of Random Lasers

There are various characteristic measurements for random lasers^[66]:

- Gain length (i.e., the distance traveled by photon over which the intensity is amplified by a factor of e),
- Average path length (i.e., the average distance of the photon travels in the medium),
- Scattering length (i.e., transport and scattering mean free path), and
- Sample size.

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Typically, when a coherent random laser is excited below its threshold, a broad spontaneous emission is observed. As pump intensity increases beyond its laser threshold, lasing peaks emerge ^[66,85]. When the pumping intensity further increases, the number of lasing modes increases and it will reach saturation at high pumping intensity ^[86,87]. The increase in lasing modes is due to the high excitation, which allows those higher losses cavities to fulfill their gain conditions. At high pumping power, laser modes begin to overlap spatially with each other and compete for gain and population inversion, as a result, only the strongest mode survives ^[88]. Also, the lasing threshold and number of lasing modes show a strong correlation with scattering length ^[66].

Photon statistic is among the most important behavior of laser radiation. For single mode coherent light, the photon number distribution is Poissonian and for single mode chaotic light, the statistic follows Bose-Einstein distribution. The photon statistic for ZnO was studied in detail using a streak camera attached to the output port of a spectrometer ^[89]. The result shown the distribution of photon changed continuously from Bose-Einstein statistics at the threshold to the Poisson statistic well above the threshold. This demonstrated that the emission from ZnO transformed from chaotic-like emission to coherent-like emission at the point of threshold, a clear evident for laser emission.

Besides, emission linewidth, frequency, polarization and directionality of emission are as important. Many experiments and theoretical models have been

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developed for such studies and the following shows some of the parameters that affect these properties:

The amount of scatterers presented affects the optical scattering path length and the transport mean free path. The higher the degree of disorder in the medium, the shorter the transport mean free path, as more scatterings are likely to occur. This enhances the amplification of light and result in a lower laser threshold [90,91]. Also, the number of modes increases with the amount of the scatterers (i.e., caused by the increases of the random cavities created) [92]. On the other hand, from the quantum theory point of view, a low concentration of particles will lead to a poor confinement of light and large overlapping of lasing modes [93].

The geometrical properties of the excitation area affect the emission of random laser as well. Report shows that stripe-like excitation area has a better chance in exciting the lasing modes [94]. Also, as the excitation area increases, laser threshold reduces [95] and the number of lasing modes increases since the possibility of having more closed loop paths in a larger excitation area is higher [91,96,97]. Furthermore, since the medium is disordered, the laser emission wavelength may various when the location of the excitation area changes [98].

It is also found that the sample size affects the laser threshold. The laser threshold generally reduces as the sample size increases. It has been suggested that this may cause by the increase in the probability of having high-quality

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cavities rather than the formation of larger cavities ^[86].

2.2.2.4 Applications of Random Lasers

Although random lasers still suffer from multi-mode emission and high laser threshold, the theoretical studies on random lasers have provided new insights on the physics of wave transport and localization. Also, it provides a feasible platform for a better understanding in chaotic theory and it has been applied in astronomy studies such as galaxy masers and stellar lasers ^[99].

The narrow-linewidth emission of random lasers is suitable for producing wavelength domain photonic codes ^[100]. It has also proposed for the encoding of documents and credit cards ^[101]. For long-range applications, random lasers have found their usefulness in military applications. Special laser dyes and scatterers were proposed to integrate with military uniforms as a form of field identification of friend and foe ^[102]. Similar methods were suggested for anti-counterfeiting as well ^[103].

For optical communication, researchers have proposed random laser as an alternative for conventional laser, especially for short wavelength application where effective mirrors are limited ^[104]. Furthermore, since the emission wavelength is determined only by the gain spectrum and should not be sensitivity to vibrations, it is proposed as a highly stable optical frequency standard ^[105].

In medical application, random lasers have also found possible applications for

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cancer diagnosis. Research has shown that cancerous tissues exhibit more random lasing peaks when they are soaked in Rhodamine 6G laser dye. Also, the average power Fourier transform (PFT) of the random laser emission spectrum reveals that the random resonator for healthy and cancerous tissues from the same organ is different ^[106]. Same analogue is applied on biomedical imaging as well, different tissues and bone types have different optical structure and mean free path, therefore, techniques such as photon density analysis and diffusing wave analysis, which based on the theory in random lasers, have proved their value in charactering tissues and monitoring blood flow during treatment and surgery ^[107].

2.3 Summary

In this chapter, we have discussed some of the intrinsic properties of ZnO. ZnO has a large bandgap of 3.4eV and a large exciton binding energy of 60 meV, which is favorable as a candidate for room-temperature UV laser. Yet, the nature of its crystal structure has given some difficulties to fabricate conventional ZnO laser for commercial industries. Therefore, an alternate method by fabricating ZnO Random laser has discussed, with its fundamental properties and working principles included briefly.

Although various applications have been proposed, ZnO random lasers still suffer from high loss and multimode operations that limit its full potential. Therefore, our objective in this thesis is to focus on the improvement of the efficiency in ZnO random laser and to have a better control over the “randomness” in this random laser.

Chapter 3

FABRICATION OF METAL OXIDE THIN FILMS

There are various methods in fabricating ZnO thin film, such as Chemical Vapor Depositions (CVD) ^[108, 109, 110, 111], sputtering techniques ^[112, 113], Molecular Beam Epitaxy (MBE) ^[114], Pulse Laser Deposition (PLD) ^[115], as well as sol- gel technique ^[116]. Each of these fabrication technologies has its individual unique advantages and disadvantages. There are several considerations for the choice of the deposition method in our research. Firstly, we would like our device to be integrable with other silicon- based device and since ZnO has a different crystal structure with silicon ^[117], it is necessary for our deposition method to be capable of depositing on lattice mismatch substrate at low temperature. Secondly, due to the stringent demand of the device's thickness, the system must be able to control the thickness of the deposition with minimum modification. Lastly, the method should be simple and cost effective for large-scale production.

3.1 Vacuum Arc Deposition Techniques

In this research, vacuum arc deposition technique ^[118] is proposed for the grow of ZnO thin films because ^[119,120].

- 1) The requirement for substrate temperature is low ($< 400^{\circ}\text{C}$),
- 2) The fabrication of ZnO thin films is not restricted with lattice-matching substrate and it is possible for large substrate production,
- 3) The microscopic particulate contains is relatively low, and
- 4) The ZnO thin films can be mass-produced at a lower cost.
- 5) Ion energy can be controlled easily and therefore the properties of the deposited film can be easily tuned.

The technique for vacuum arc growth ZnO thin films are well established ^[121,122] and results have shown promising properties for potential applications in field emission displays ^[123], and solar cells ^[124]. Also, ZnO UV optical devices, such as optical waveguides, lasers and light-emitting diodes have been reported recently ^[125,126,127].

Figure 3-1 Schematic of a typical vacuum arc deposition system

Figure 3-1 shows the schematic diagram of a typical vacuum arc deposition system. This system comprises of a deposition chamber, a target chamber and a filter. The film deposition is processed in a vacuum environment, evacuated through a vacuum pump. The substrate is placed on a substrate holder and located inside the deposition chamber. Different types of process gases are supplied through the gas inlets at the deposition chamber.

The deposition is initiated by igniting an arc at the target within the target chamber and plasma is produced as a result of this arcing. Afterward, this plasma is being guided along the filter by a magnetic field and deposit onto the substrate at the deposition chamber.

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To improve the quality of the deposited film, a heating element is usually placed in close vicinity to the substrate holder to control the substrate temperature ^[128]. To reduce the macroparticle contamination in the deposited films, a biasing supply ^[129] and shielding ^[130] is commonly applied. The biasing supply is connected to the substrate holder whereas the shield is normally located at some distance away from the substrate, to avoid the substrate to be in line-of-sight with the target. To reduce the macroparticle contamination further, macroparticle filter ^[131,132] has also been used in the deposition process. This filter is usually found in between the deposition and target chamber.

The electrical terminal of the target chamber can be connected in two ways. The target can be connected to negative or positive terminal of the vacuum arc supply and with the other terminal connect to a reference ground. The deposition with the former connection is known as the cathodic arc deposition technique ^[133], whereas the latter is known as the anodic arc deposition technique ^[134].

The vacuum arc power supply can either be in a continuous ^[135] or pulse ^[136] operation. For continuous operation mode, the thickness of the deposited film can be controlled by adjusting the exposure time of the substrate for the plasma. This can be done by installing a shutter inside the target chamber. It is also possible to control the thickness of the film by controlling the amount of arcing pulse in the pulse operation mode, but precise calibration and extremely stable electrical arcing supply is required.

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Cooling system is extremely important in vacuum arc deposition technique. Due to the large amount of arcing current flowing through the target material, tremendous heat can be generated and this will result in the melting of the target material. Besides, the overheated target will also result in a higher macroparticles generation and contaminate the deposited sample ^[137].

3.1.1 Macroparticle Control

Without a proper control of macroparticles, the deposited thin film is usually contaminated badly and unsuitable for device fabrication. Therefore, macroparticle control is one of the most important factors that should not be neglected in vacuum arc deposition system.

Macroparticles are generated from the non-stationary cathode spots. When the arc is triggered in the target chamber, the arc current is usually being concentrated within a small number of discrete sites called cathode spots. Plasma is formed as a result from these cathode spots, but unwanted macroparticles also being produced due to the high energy.

To reduce the macroparticle contamination, different modes of arcing operations have been proposed ^[138,139]. However, this is not preferred for applications where the generation of ions with high kinetic energies is crucial ^[140]. Hence, significant interests have been focused to reduce the contamination of macroparticles in the vacuum arc deposition technique.

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Various methods for macroparticles reduction have been studied, such as field coils induced rapid cathode spot motion ^[141] and the lowering of the target temperature by the reduction of the arc current ^[142], however, the latter seems to be counterproductive with the deposition rate. Besides, negative biasing of the substrate ^[129] and the manipulation of the background pressure ^[143] have been demonstrated to lessen the macroparticle contamination. Steered and shielded arc deposition methods ^[144,145] have also been studied. In steered arc deposition technique, a magnetic field is installed to guide the motion of the arc spot and for the shielded arc deposition method, a shield is placed in between the substrate and the target, along the path of the plasma.

Lastly, macroparticles contamination can also be reduced with the conventional filtering method, such as dome filter ^[146], classical 90-duct filter ^[147], 45°-duct filter ^[148], S-duct filter ^[149], rectilinear filter ^[150], freestanding magnetic coil filter ^[151], stroboscopic filter ^[152], Venetian blind filter ^[153] and rotating blade filter ^[154]. Among them, the more successful ones are the magnetic filters ^[155].

Figure 3-2 Schematic of the off-plane double-bend filter

Hence, magnetic filtering is used in this research. In this method, the plasma ions are guided by the magnetic field within the filtering-duct. Figure 3-2 shows a schematic diagram of an off-plane double-bend (OPDB) filter^[156]. This filter has two-torus bends at 90° and 45° with respect to the axis of the cathodic vacuum arc source^[157,158]. Electromagnetic coils are built in the exterior wall of the filtering duct and magnetic field is induced within the filtering duct, parallel to the axis of the filter. This magnetic field will guide the charged plasma towards the deposition chamber in a spiral motion due to the Lorentz force produced by the magnetic field, the charge and the velocity of the ions. The neutrally charged macroparticle is therefore unguided and being captured or reflected by the baffles located in the interior wall of the filtering duct. The OPDB filter is effective in eliminating macroparticle contamination in the

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deposited films with large number of macroparticles hitting the walls of the filtering duct ^[156]. Also, the structure of the filter is designed to have the optimum output efficiency while keeping the losses of plasma in the filter to a low level ^[156].

3.2 Filtered Cathodic Vacuum Arc Deposition Technique

Figure 3-3 Schematic of the FCVA system

Figure 3-3 ^[159] shows the schematic diagram of a typical Filter Cathodic Vacuum Arc (FCVA) system for the deposition of our metal-oxide thin films. Metal with high purity (99.9%) is used as the target material and it is connected to the cathode of a vacuum arc continuous biasing supply. Metal target is preferred as compared with metal oxide target because:

- The cost of metal target is normally more economical than a compound target.
- Metal target usually has a lower melting point in which, a lower biasing voltage is required to trigger the arc.

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- Metal target has better conductivity and hence, better arcing sustainability.

As for the preference of biasing method, cathodic arc deposition is selected because:

- A higher percentage of the material evaporated from the target can be ionized ^[160],
- ions exist as multiple charge states in the plasma ^[161], and
- the kinetic energies of the ions generated are higher ^[162].

Furthermore, continuous biasing operation is preferred over pulsed operation because it is more suitable for high quality, large-areas thick films fabrication ^[163].

To prevent the metal target from being overheated and melted in the deposition process, cooling water is being circulated in close proximity with the base of the target. Apart from melting, overheating of the target will also result in the increase in generation of macroparticles ^[164].

Arcing is generated when the trigger strikes upon the target surface. It is self-sustained and confined within the magnetic field generated from the cathode coil. The arc spot is restricted within the target surface by a small gap in between the target and the isolating shield. Metal plasma is generated in this process and the magnetic field from the magnetic filtering-duct will guide the

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plasma towards the deposition chamber. This metal plasma will react with the injected oxygen gas and form metal oxide before it arrives onto the substrate.

3.3 Twin Arms FCVA System

To fabricate multilayer structure, a customized FCVA system is specially designed and built, as shown in Figure 3-4. This system comprises of an addition arm, which includes a magnetic filtering duct and a target chamber. As such, alternate deposition of two different metal oxides within a single vacuum process is possible.

Figure 3-4 Photograph of the twin arms filter cathodic vacuum arc system

To improve the quality of the thin film and also, increase the precision of thickness control, various troubleshooting, modifications and optimization were performed.

3.3.1 Cooling System

As mentioned earlier, cooling system is essential and it is extremely important for FCVA deposition. Proper cooling system can prevent target from overheating and also, reduces the amount of macroparticles contamination. Various problems arise and following addresses some of the countermeasures:

It was found that any large fluctuation of the target temperature affects the stability of the arc, hence, additional chillers were installed to maintain the temperature of individual target.

Typical FCVA system uses indirect target cooling method, in which, the base of the metal target is bonded onto a target holder with Indium pallet and the heat from the target is exchanged with the target holder before loses to the cooling water, as shown in figure 3-5(a). Hence, the efficiency of indirect cooling method is poor and also, due to the low melting point of the bonding pallet, this method is not suitable for prolong deposition. In our deposition system, a direct target cooling method is explored.

For direct cooling, as shown in figure 3-5(b), the target for our system is specifically manufactured to the dimension of the target holder, attached onto the cooling system and allows a direct contact between the base of the target material and the cooling water. This method improves the cooling efficiency and lengthens the allowable duration of the deposition process by double.

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The drawback for this direct cooling method is that, the sealing of the cooling water from the vacuum chamber is dependent on individual target and any defects at the base of the target will result in a water leakage. Also, because the base of the target is in direct contact with the cooling water, a steep thermal gradient is present across the target whenever there is arcing on the surface of the target. These thermal shocks and corrosion from the cooling water causes hairline cracks and leads to minor water leakage, which can only be observed with a slight increase in the chamber vacuum pressure. Although the leakage is minor, it affects the quality of the deposited thin film and will also lead to severe damage to the system.

The direct cooling method is improved by installing a copper plate in between the metal target and the exchange of the cooling water, as shown in figure 3-5 (c). It acts as a buffer layer to protect the base of the target and also, reduces the steepness of the thermal gradient across the target. Copper is chosen because it has a higher corrosion resistance as compared with the metal targets and it is also a good conductor of heat. Various thickness of copper plate is tested and 1 mm thickness is chosen for the balance of the damage protection and also without much compromise to the cooling efficiency.

Figure 3-5 Schematic of the target assembly with (a) indirect cooling method, (b) direct cooling method and (c) improved direct cooling method

3.3.2 Plasma alignment

Due to its charged nature, plasma can be guided with the present of an external magnetic field. In the FCVA system, guiding field is generated by the magnetic coils located on the exterior wall of the magnetic filtering duct. When a direct voltage is applied across the coil, a magnetic field will be induced in the direction perpendicular to the close loop of the current coil as governed by the Ampere's law.

With the charge (q), the velocity of the moving ion (v), electrical field (E) and the external magnetic field (B), a continuous force will be acting on the ions at the direction perpendicular to the direction of the ion velocity and the external magnetic field, which is known as the Lorentz force (3.1). This force will guide the ions to travel in a spiral motion along the filtering duct axis and reach the deposition chamber.

$$F = q(E + v \times B) \quad (3.1)$$

In order to have a better control on the 1) direction of the plasma beam, 2) growth rate and 3) uniformity of the deposited films, an additional scanning coil is installed at the end of the filtering duct, as shown in figure 3-5. This scanning coil is made of 2 sets of perpendicular magnetic coils in which, their magnetic fields are vertically and horizontally perpendicular to the direction of the plasma beam. By adjusting the strength of these magnetic fields, the diameter of the plasma beam can be tuned; also, the direction of the beam can be steered for alignment.

Figure 3-6 Photograph of the scanning coil

The initial deposition rate was low because in a typical FCVA system, the magnetic field and hence, the plasma beam diverged as it leaves the guiding coil, as shown in figure 3-6. The simulated result is shown in figure 3-7 and the brightness of the figure is proportional to the strength of the magnetic field. The strength of the field is strongest at both ends of the magnetic coil and diverged as it leaves the coil.

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Figure 3-7 Schematic of the magnetic field within the FCVA system

Figure 3-8 Simulated result of the magnetic field within a typical FCVA system

To reduce the spread of the plasma beam, an additional magnetic coil is built at the outer wall of the substrate holder, as shown in figure 3-8.

Figure 3-9 Photograph of the additional magnetic coil and its DC power supply

By ensuring the distance between the 2 magnetic coils is shorter than the length of the filtering duct, most of the magnetic field can be coupled between these 2 coils. Thus, the new-coupled magnetic field will be redirected through the substrate holder as shown in figure 3-9, and hence, the plasma. Also, the focus of the plasma will be maintained. To further enhance the magnetic field at the substrate holder, a soft iron plate, with relatively permeability of ~ 200 is placed at the base of the substrate holder. The simulation result of the redirected magnetic field is shown in figure 3-10, it shows that the field is now focused and directed towards the substrate holder, before it returns to the magnetic filter. The magnetic field at various locations has been measured with a gauss-meter and it has been confirmed that the redirected field has strengthened by 5 times. This modification improves the growth rate by 3 times and yet, maintaining the quality of the deposited films.

Figure 3-10 Schematic of the magnetic field within the modified FCVA system

Figure 3-11 Simulated result of the magnetic field within the modified FCVA system

3.3.3 Deposition thickness control

In most of the multilayer optical structures, the thickness of the layer is crucial as it governs most of the optical properties, from exciton transition energy to lasing emission ^[165]. Various methods to improve the control of deposition thickness were tested for our FCVA system.

A modified Faraday cup method ^[166] was first proposed to measure the thickness of the deposited film. Faraday cup is a conductive cone that is designed to collect charged particles in vacuum. When a packet of ions collided with the conductive cup, the cup will have a gain in a small net charge while the ions are being neutralized. The gained charges will be discharged in a form of current and the magnitude of this small current is equivalent to the number of impinging ions. The number of charges hitting the cup can then be determined by the magnitude of this discharge current.

In our system, the inner surface of the metallic filtering duct is used as the “Faraday metal cup”. Ideally, the amount of ions captured by the metallic duct will be proportional to the un-captured ions that arrive onto the substrate. But in reality, the accuracy of the measuring method is in the range of micrometers. Firstly, whenever there is a current flows, the coil will be heated up, which increases the electrical resistance of the magnetic coil. This causes a fluctuation in the supply current and affects the magnetic field for plasma guiding. This creates a disturbance to the proportion of ions capturing by the filtering duct and the ions that arrive onto the substrate and hence, causes error to the

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thickness measurement. Secondly, the discharge current measurement at the filter duct has a resolution of 0.1 mA, hence, the uncertainty with the number of ions measured will be in a range of 10^{16} if we assumed that the average ion has a charge of $1.6 \times 10^{-19} \text{C}$. Lastly, the delay in the indirect data processing further degrades the accuracy.

An alternative based on crystal oscillation was tested. This crystal oscillator relies on the piezoelectric effect. A piezoelectric material vibrates whenever an external electric field is presented, and its vibration frequency is proportional to the mass of the crystal. When material is deposited on top of the crystal, its vibration frequency will be changed due to the overall change in mass. By measuring the difference in frequency, the thickness of the material grown can be deduced.

This thickness monitor includes a controller at figure 3-11, a signal conversion circuit at figure 3-12 and a crystal holder at figure 3-13.

Figure 3-12 Photograph of the thickness control monitor

Figure 3-13 Photograph of the signal conversion circuit

Figure 3-14 Photograph of the crystal holder

Due to the presence of the large amount of noises, the thickness monitor did not function properly when it was first installed. The noise was found to be originated from the magnetic field for plasma guiding and the discharging current from the plasma ions when they collide with the crystal holder. Hence, magnetic shielding is designed and incorporated into the system.

This magnetic shield is made of a soft iron pipe, as shown in figure 3-14. Soft iron is chosen because of its high relative permeability. As we all know, reluctance ($\mathcal{R}$) is a function that is inversely proportional to the cross sectional area (A) and the material permeability (μ) (3.2).

$$\mathcal{R} = \frac{l}{\mu A} \quad (3.2)$$

Figure 3-15 Photograph of the soft iron pipe for magnetic shielding

Hence, by placing a lower reluctance material along the magnetic field, majority of the flux will be diverted through the low reluctance path. This will guide the magnetic field to travel around the soft iron pipe and bypass the crystal holder. The simulation of the diverted magnetic field is shown in figure

3-15. Also, this will act as a physical shield that prevents the plasma from colliding with the crystal holder and generate any unnecessary discharge current. The installed crystal holder with its magnetic shielding is shown in figure 3-16.

Figure 3-16 Simulated result of the diverted magnetic field at the cross section of the magnetic shield

Figure 3-17 Photograph of the crystal holder with magnetic shield installed

3.3.4 Optimization of ZnO growth

Since ZnO will be used as the gain medium for most of our optical device, it is important to optimize the deposition process before embarking for a more complex structure. The efficiency of the light emission is chosen as the benchmark for our optimization. The various perimeters that can be fine-tuned are; the plasma energy, the oxygen contains and also the substrate temperature. Different combinations of plasma energy and oxygen injections were tested and compared. We found that for our system, the best quality of ZnO was obtained with an arcing voltage of 70V and with the flow rate of the oxygen at 170 sccm, as shown in figure 3-17.

Figure 3-18 Plot of the PL emission intensity vs oxygen flow rates during deposition process

We discovered that although a higher plasma energy may improves the interaction between Zn plasma and the injected oxygen, the high arcing current will also increase the marcoparticles generation and therefore degrades the quality of the thin film. As for the injection of oxygen, we discovered that when

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the oxygen flow rate is set below 165 sccm, the optical emission of ZnO is relatively poor, due to the zinc rich nature of the ZnO film. As for the flow rate beyond 180 sccm, we noticed another decrease in term of optical emission intensity as in figure 3-18. This is because the excess supply of oxygen increases the chamber pressure; therefore, shortened the collision time of the Zn plasma and loses its energy in the form of scattering.

Figure 3-19 Plot of the deposition rate vs oxygen flow rates

3.3.5 Optimization of Al_2O_3 growth

Al_2O_3 has been chosen as one of the metal oxide in the DBR design because of its optical transparency at ZnO absorption and emission band, and also its high refractive index contrast with ZnO. Therefore, the benchmark for Al_2O_3 is selected for the optical transparency at 350nm and above. While in the process of optimization, we have noticed that the Al plasma is extremely difficult to create and sustain, which was found to be caused by the insulating nature of Al_2O_3 . In the process of deposition, a layer of Al_2O_3 is formed on the surface of the aluminum target. This layer increases the resistance of the surface and as a result, the arc extinct. This extinction of arc could be overcome by increasing the arc current from 70 to 160 A.

However, by having a large amount of arcing current, the surface of the aluminum target melts within the first minute of the deposition. This melted surface causes the trigger to be stuck onto the target and result in a system failure. Hence, the trigger assembly was modified and additional manual control mechanism was installed, as show in figure 3-19 and 3-20.

Figure 3-20 Photograph of the trigger mechanism before modification

Figure 3-21 Photograph of the trigger mechanism after modification

Upon the modification of the striking assembly, Al_2O_3 film has successfully grown on the UV graded quartz substrate and optical transmission at UV range is measured. We have discovered that as the oxygen flow rate increases to 150 sccm and more, the optical transmission is beyond 99% and therefore, we have chosen to operate at 150 sccm for the best balance between the optical transmission and the deposition timing.

3.4 Summary

In this chapter, a brief introduction of the FCVA deposition system is given, including various filtering methods to improve the quality of the deposition and also, its working principle. For this research, a twin arms FCVA, which is capable of depositing two different types of metal oxide within a single process was designed and built. Several engineering challenges, such as difficulties associated with cooling, plasma alignments and thickness control were elaborated and solutions were provided. Lastly, the optimizations of the metal oxides (ZnO and Al₂O₃) were also discussed in detail.

Chapter 4

LOW TEMPERATURE PHOTOLUMINESCENCE OF ZnO RANDOM LASERS

4.1 Low Temperature Photoluminescence Setup

In this project, photoluminescence (PL) spectroscopy ^[167] is employed as our main characterization technique to study the optical properties of our lasing devices. When electron in the optical medium is excited by light, photon will be absorbed. The excited electrons will transit to a higher permissible excited states and return back to the lower states by radiating photons (radiative recombination ^[168]) or phonons (non-radiative recombination^[168]). PL spectroscopy refers to the measurement of this radiative recombination process. The energy of the emitted light relates to the difference in energy levels between the states of electron involved in the transition, which can be used to deduce the recombination processes. By studying the emission spectrum, various radiation characteristics can be

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determined – the band gap energy, presences of impurity and defects, exciton energy ^[169], and also some of the laser characteristics ^[170].

For our characterization, a low temperature PL system is built and the schematic layout is shown in figure 4-1.

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Figure 4-1 Schematic layout of the optical setup for the low temperature photoluminescence system

The setup consists of a Nd:YAG laser (Surelite III-10Hz), a Surelite Separation

Package (SSP), a cryostat, a monochromator (Oriel 77200 1/4m grating), a He:Ne laser, lenses and mirrors. They were aligned at the same optical height with respect to the optical table and secured. A photograph of our low temperature PL setup is shown in figure 4-2.

Figure 4-2 Photograph of the low temperature photoluminescence system

The laser in our system has a fundamental wavelength at 1064nm. It can generate lasing wavelength of 532, 355 and 266 nm through a combination of different harmonic crystals. It has a repetition rate of 10Hz and a pulse width of 120 ps. The maximum output average energy is 80 and 25 mJ for the emission at 355 and 266 nm respectively.

For PL studies, the stability of the laser output power is important because our setup measures the average of 10 excitations per wavelength reading.

Therefore, extra emphasis is given for the stability of our laser before the rest of the optical alignment.

Our laser operates at pulse mode through Q-switching^[171] and controlled by an Acousto-Optic Modulator (AOM)^[172]. AOM relies on the acousto-optic effects, in which, sound waves or radio waves can be used to create diffraction in light. It is an effect that changes the material permittivity with respect to the applied mechanical strain, caused by an external wave. The laser was unstable at first because this AOM is extremely sensitivity to the ambient temperature. With a large input range of few tens kHz, It was tuned at an increment of 10 Hz till its output pulse was stabilized and its output power was consistence. The output power and the repetition rate were measured with an oscilloscope and a power meter.

To produce a uniform laser beam profile, the optics within the laser were realigned. Misaligned optics causes poor beam profile for sample excitation and also, damages the laser. The uneven distribution of laser intensity in a non-uniform beam creates an undesired nonlinear self focusing effect^[170] across the Nd:YAG rod and harmonic crystals, leading to uneven thermal stress distributed across the crystal and damage the crystal eventually. The quality of the beam was adjusted by tuning the mirror located at the end of the cavity. Due to the infrared natural of the fundamental beam, laser alignment paper, a special paper that reacts sensitively with laser intensity was used.

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UV emission from our Nd:YAG laser are generated with the harmonic crystals. It is a nonlinear optical process ^[171], where photons interact with each other within an optically anisotropic material. It relies on the phase matching of the momentum or in another word, the matching of the refractive index along the ordinary and extraordinary axis of an anisotropic material. As such, harmonic generation can only be seen at a particular angle.

There are 2 types of second harmonic crystal (SHG), in Type I SHG, two photons having ordinary polarization with respect to the crystal will combine to form one photon with double the frequency and extraordinary polarization. In Type II SHG, two photons having orthogonal polarization will combine to form one photon with double the frequency and extraordinary polarization. For our system, Type I is used for 3rd harmonic generation and Type II is used for the 4th harmonic generation.

The alignment of the harmonic crystal was performed with the help of the laser linewidth bandpass filter and a power meter. The angle of the harmonic crystal was tuned in a change of 0.1 degree until its output intensity reaches its maximum.

At the output of our laser, a pair of dichroic mirrors was installed. Its purpose is to filter the unwanted 1064 and 532 nm, and only allows the 355 nm laser to transmit. This is to reduce the unwanted components from pumping and prevent unnecessary heating of the sample under investigation.

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The alignment of optics was performed with the He:Ne laser. It was aligned with the objective lens in front of the input slit assembly, passing through the cryostat without the sample. Position of the objective lens was adjusted until the detector output signal registered a maximum. This step was repeated until the input and output slit widths reduces to 10 μm .

For PL characterization, the sample is placed on a copper holder, within a close-circuit cryostat. Liquid nitrogen is used as the heat exchange agent in our cyro-system. It has the capability of reaching 77 K and maintains for 2 hours. The sample is insulated in vacuum of 10^{-3} torr from the external heat, through a rotary pump. An external temperature controller is used to control the temperature of the sample holder. During the characterization process, the excitation laser source passes through a linewidth filter, an optical attenuator and focused by a cylindrical lens before it arrives onto the sample. The filter is used to refine the excitation source and the optical attenuator is used to provide a more define adjustment for its power. Cylindrical lens is preferred because it allows gain measurement through variable line strip method. The light emission from the sample is focused into a parallel beam by an objective lens at its working distance. The emission lights is diffracted by the grating, inside the monochrometer and detected by the photomultiplier tube (PMT). To improve the signal to noise ratio, a lock-in amplifier is used to filter the unwanted white noise before the data capturing process by a Labview program.

4.2 Low Temperature Random Lasing studies in ZnO thin film

Since the realization of random laser in our FCVA deposited ZnO thin film, many of its optical properties have been studied, such as the ASE characteristics^[173], random lasing actions^[174], various recombination processes^[175], and also high temperature random lasing behaviors^[176]. To have a more complete and a better understanding of the fundamental ZnO random lasing behavior, low temperature photoluminescence characterization is investigated on the ZnO thin film. The measurement is taken from 77 K to room temperature. Our results show that as the temperature reduces, the laser emission blue-shifted. Moreover, the laser threshold decreases and the FWHM narrowed with respects to the temperature drops. We also observed the transformation of the emission process as the temperature increases.

In this experiment, ZnO thin film is fabricated with our twin arms FCVA system. The thin film is deposited on UV grade quartz substrate at 600 K, with a film thickness of 200 nm. The oxygen flow rate is 170 sccm and the substrate holder is rotated at 30 rev/min to ensure the uniformity of the surface. The film is treated with post annealing at 1100 K to improve the size of the grains and therefore the random lasing properties.

Figure 4-3 shows the XRD pattern of our ZnO film. It can be seen that the film possess a polycrystalline Wurtzite structure with $2\theta = 34.7^\circ$ (002) preferred orientation. The SEM image of our thin film is shown in Figure 4-4. Clearly,

boundaries can be seen with an average diameter of~ 80nm, which are responsible for the scattering feedback in random lasing.

Figure 4-3. XRD spectra of FCVA deposited ZnO thin film at 600 K

Figure 4-4. SEM image of the FCVA deposited ZnO thin film at 600 K

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To study the low temperature photoluminescence properties, our sample is placed at a cryostat and cool to 77 K with liquid nitrogen. Temperature dependent photoluminescence measurements have recorded in the range of 77 to 300 K. The sample was excited with a Nd:YAG laser, operated at 355 nm and the sample radiation was measured with a photomultiplier tube, attached to a monochromator.

Figure 4-5(a) shows the ZnO photoluminescence from 80 to 300 K at an excitation of $4\text{MW}/\text{cm}^2$ and figure 4-6(a) shows the radiation excited at $5\text{MW}/\text{cm}^2$. We can see clearly that the emission wavelength is blue-shifted from 390 to 377 nm as the temperature reduces, which is corresponding to 3.17 and 3.27 eV respectively. Also, the emission intensity increases about 10 times at 77 K when it is compared with the radiation intensity at 300 K, in which, it is more obvious at higher excitation power in figure 4-6(a). The relationship between emission intensity, emission wavelength, and sample temperature is shown in figure 4-5(b) and figure 4-6(b).

Figure 4-5 (a) Emission spectrum measured perpendicularly with the surface of the ZnO thin film at various temperatures and excited at 4 MW/cm². (b) Shows the relation between the emission wavelength, emission intensity and the sample temperature.

Figure 4-6 (a) Emission spectrum measured perpendicularly with the surface of the ZnO thin film at various temperatures and excited at 5 MW/cm². (b) Shows the relation between the emission wavelength, emission intensity and the sample temperature.

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Figure 4-7 shows a typical light- light curve at various temperatures. The present of a “kink” is a signature of laser threshold and occurs when the gain is equal to its loss. Together with the multiple lasing peaks observed at figure 4-5 and figure 4-6, we can conclude that the lasing action in our ZnO thin film is due to the random feedback path created by the grain boundaries. This is because our sample does not have any Fabry-Perot structure to sustain conventional lasing. We also discovered that the laser threshold reduces from about 0.37 MW/cm² to 0.30MW/cm² when the temperature of the sample decreases from room temperature to 80 K.

Figure 4-7 Light-light curves of the polycrystalline ZnO thin films at various temperatures.

To compare the change in FWHM with respect to sample temperature, various emission spectra at different temperatures and different excitation power is

shown in figure 4-8.

Figure 4-8 Emission spectrum measured perpendicularly with the surface of the ZnO thin film at various temperatures and excited intensities, the arrow indicates the red-shift of the emission peak.

It is observed that as the sample temperature increases from 80 to 300 K, the FWHM increases from 2.7 to 4.8 nm. We also noticed the red-shifting of the peak intensity (as indicated by the black arrow) with respect to the excitation power is more obvious in the higher temperature as compared with 80k at which, the red-shifting is almost unnoticeable. Furthermore, at higher temperature, the lineshape of the spectrum is asymmetric with a low energy tail, which shows the characteristic of a degenerate plasma^[177].

Figure 4-9 shows the summary of our findings. It shows the relations between peak wavelength, peak intensity, laser threshold, FWHM, and the red-shifted peak intensity with respect to the change in the sample temperature.

Figure 4-9 Summary of the change in peak wavelength, peak intensity, laser threshold, FWHM, and the red shifted peak intensity with respect to the change in sample temperature

As the temperature reduces, the wavelength of the emission peak increases from 390 to 380 nm rapidly and slows down as it approaches 380 nm. By taking the temperature-boarding effects into account, reported articles have suggested that this temperature dependent emission is corresponding to the second LO-phonon replica of the A-free exciton^[178,179]. This is slightly different from most of the reported ZnO low temperature PL emission in which it is usually

dominated by the near band edge emission of the radiative decay of donor bound exciton B ^[180,181,182,183]. Probably, this is because our excitation source has a lower energy and nearer to the band edge of ZnO at 355nm, in which, will be less favorable for the donor-bound exciton B emission.

It is also found that the emission intensity reduces with respect to the increase in temperature and the laser threshold of the ZnO thin film increases exponential with temperature. The relevant dependence can be fitted by the following equation,

$$I_{th}(T) = I_o \exp\left(\frac{T}{T_o}\right) \quad (4.1)$$

Where I_o is a constant, T is the measurement temperature, $I_{th}(T)$ is the threshold intensity, T_o is the characteristic temperature and was found to be ~90 K. This can be explained by the broadening of the quasi-fermi function of electrons and holes at elevated temperature ^[184]. Therefore, the exciton enhancement effect reduces and results in a higher threshold ^[185].

We also observe the widening of the FWHM and the increase in red-shifting of the peak emission correspond to the increase of excitation intensity as the temperature increases. Together with the asymmetric emission linewidth at high excitation intensity, report has been suggested that this temperature dependent emission mechanism is a result of the transition from exciton-exciton scattering process to the formation of electron hole plasma (EHP) ^[186].

It is known that the transition from an exciton gas to an EHP is in a continuous

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form in direct bandgap semiconductor due to the short lifetime of the carrier pairs, which prevent the evolution of the phase separation below a critical temperature predicted by quasi-equilibrium thermodynamics.^[187]

As the temperature increases, the homogeneous width of the exciton becomes comparable to its binding energy and therefore, leading to the formation of the EHP^[186]. The laser threshold at room temperature is also reported as a value close to the Mott density of ZnO^[187]. This suggested that the condition at room temperature is highly favorable for EHP emission.

The red-shifting of the emission spectrum is a distinctive signature of the Band Gap Renormalization (BGR) from EHP. In a typical EHP, due to Pauli principle, two parallel spin electrons are forbidden to sit at the same unit cell. As the density increases, the exchange energy increases the average distance between the electrons with parallel spin and consequently reduces their total repulsive Coulomb energy^[187]. This reduction in energy will mean a lowering of the total energy of the electron system and result in the red-shift in the EHP spectrum as the excitation power increases.

4.3 Summary

A low temperature photoluminescence setup was assembled for the characterization of the ZnO optical emission and various optimizations were discussed. Our setup was used to investigate the low-temperature random laser behavior of the FCVA deposited ZnO thin film. The temperature dependent of laser threshold, emission spectrum width and the red shifting of the emission were observed. Also, the continuous transformation from exciton -2LO recombination process to EHP recombination process was noticed to be more prominent as the temperature increases.

Chapter 5

ANALYSIS AND DESIGN OF DISTRIBUTED BRAGG REFLECTOR AND VERTICAL-CAVITY SURFACE- EMITTING LASERS

To optimize the optical properties of our multilayer light emitting device, simulation studies are essential. In this chapter, a model based on transfer matrix method is developed to study the optical behaviors of distributed Bragg reflectors. This will be used to investigate the anti-resonant reflecting optical waveguide (ARROW) vertical-cavity surface-emitting lasers (VCSELs), a complex laser device operating at the above threshold condition.

5.1 Distributed Bragg Reflector

Distributed Bragg reflector (DBR) is a periodic structure, made of multiple

pairs of different refractive index dielectric materials. When light propagates through a composite structure that made of two different dielectric mediums, it will be reflected at the junction of the discontinuity at which, the two different mediums met. The amount of light reflected at one such boundary is usually small. However, if layers of alternating dielectric materials are stacked together periodically and each layer has an optical thickness that is a quarter of the optical wavelength, Bragg condition of a particular oscillation frequency can be obtained. The reflected light from each of these boundaries will be added in phase constructively and produces a large reflection at Bragg Frequency. The main advantage of DBR as compared with metallic mirror is that, in general, DBR can produce a higher reflectivity in the UV range because most of the metal has its plasma frequency below UV.

5.1.1 DBR Model

The transfer matrix model is develeoped with reference^[188]. In figure 5-1, at the boundary of two different dielectric mediums, the tangential components of both electric and magnetic fields must be continuous such that

$$E_I = E_{II} + E_{rI} = E_{II} + E'_{rII} \quad (5.1)$$

$$E_{II} = E_{III} + E_{rII} = E_{III} \quad (5.2)$$

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Figure 5-1 The schematic diagram of the electric field between 2 dielectric boundaries

E and H fields are related through the refractive index and the unit propagation vector:

$$\vec{H} = \sqrt{\frac{\epsilon_0}{\mu_0}} n \hat{k} \times \vec{E} \quad (5.3)$$

Also, when a wave propagates through a medium, it will undergo a phase shift of $k_0 (2n \cdot d \cdot \cos \theta / 2)$ where k_0 is the wave number, n as the refractive index of the medium, d be the thickness of the medium and θ as the incident angle. This will be denoted as $k_0 h$ where $h = (2 \cdot n \cdot d \cos \theta / 2)$.

Therefore,

$$E_{ill} = E_{il} e^{-ik_0 h} \quad (5.4)$$

$$E_{rll} = E'_{rl} e^{ik_0 h} \quad (5.5)$$

With the above equations, the E and the H field propagating in 2 mediums can be resolved as follows

$$E_I = E_{II} \cos k_0 h + H_{II} (i \sin k_0 h) / Y_1 \quad (5.6)$$

$$H_I = E_{II} Y_1 i \sin k_0 h + H_{II} \cos k_0 h \quad (5.7)$$

With

$$Y_1 \equiv \sqrt{\frac{\epsilon_0}{\mu_0}} n_1 / \cos \theta_{ill} \quad (5.8)$$

Or in a matrix form;

$$\begin{bmatrix} E_I \\ H_I \end{bmatrix} = M_I \begin{bmatrix} E_{II} \\ H_{II} \end{bmatrix} \quad (5.9)$$

where

$$M_I = \begin{bmatrix} M_{11} & M_{12} \\ M_{21} & M_{22} \end{bmatrix} = \begin{bmatrix} \cos k_0 h & (i \sin k_0 h) / Y_I \\ Y_I i \sin k_0 h & \cos k_0 h \end{bmatrix} \quad (5.10)$$

In general, with p number of layers, the E and H field in the first boundary and last boundary can be related as

$$\begin{bmatrix} E_I \\ H_I \end{bmatrix} = M_I M_{II} \dots M_p \begin{bmatrix} E_{p+1} \\ H_{p+1} \end{bmatrix} \quad (5.11)$$

And the reflection coefficient r and transmission coefficient t can be deduced as:

$$r = \frac{Y_0 m_{11} + Y_0 Y_s m_{12} - m_{21} - Y_s m_{22}}{Y_0 m_{11} + Y_0 Y_s m_{12} + m_{21} + Y_s m_{22}} \quad (5.12)$$

$$t = \frac{2Y_0}{Y_0 m_{11} + Y_0 Y_s m_{12} + m_{21} + Y_s m_{22}} \quad (5.13)$$

The above equations can be resolved with Matlab. We have chosen Al_2O_3 and ZnO as our dielectric metal oxide due to the fact that there is a large difference in the refractive index between the two metal oxides, the bandgap of Al_2O_3 is larger than ZnO such that it is almost transparent to ZnO emission and also, these metal oxides are possible to grow with our FCVA system. In order to be realistic for our simulation results, we have considered the refractive index for Al_2O_3 ^[189] and ZnO ^[190] as a frequency dependent component as shown in figure 5.2 and 5.3. Also, a 5% variation in thickness has added to factor the experimental deposition errors.

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Figure 5-2 A plot of the Refractive index versus wavelength for Al_2O_3

Figure 5-3 A plot of the refractive index versus wavelength for ZnO

5.1.2 Results and discussions

Figure 5.4 shows a comparison between the simulation results with and without the consideration of frequency dependent refractive index. The location of the peak reflection is shifted by 15 nm as a result of the above consideration.

Figure 5-4 A comparison of reflection spectrum between refractive index with and without frequency dependent component.

Due to the strong contrast in the refractive index difference between ZnO and Al₂O₃, 12 pairs of ZnO and Al₂O₃ is sufficient to achieve a reflectivity of 97%. The result of the reflection spectrum against different no. of dielectric pairs is shown in figure 5-5. Figure 5-6 shows the maximum reflection against different no. of dielectric pairs.

Figure 5-5 Reflection spectrum with different no. of Dielectric pairs

Figure 5-6 Maximum reflection with different no. of Dielectric pairs

5.2 Anti-Resonant Reflecting Optical Waveguide Vertical-Cavity Surface-Emitting Lasers

While in the process of setting up our fabrication system and characterization systems, we have performed theoretical studies on a complex multilayer laser device named anti-resonant reflecting optical waveguide (ARROW) vertical-cavity surface-emitting lasers (VCSELs) with our transfer matrix model and rate equations.

VCSEL is a type of semiconductor laser in which, the emission direction is perpendicular from the top surface, contrary to the edge emission of the conventional edge-emitting semiconductor laser^[191]. Usually, the gain medium is sandwiched horizontally between 2 DBRs to provide optical feedback. There are various advantages for VCSEL as compared with the conventional laser, such as the possibility to be tested on wafer before cleavage^[192], lower divergence angle of the output beam due to its large output aperture^[193], lower threshold because DBRs have a better reflection than cleaved edge in the conventional laser^[194] and VCSEL can be easily tuned by change the thickness of the reflectors layers^[195]. However, due to their weak lateral index-guiding nature, they are very susceptible to gain spatial hole burning and thermal waveguiding, which limits the size of VCSEL for single mode operation^[196].

Therefore, ARROW structure for VCSELs has been proposed^[197] to provide strong lateral radiation losses, which are highly mode dependent, thus filtering

out higher-order spatial modes. A schematic structure of ARROW VCSELs is shown in figure 5-7. In this structure, the low-index core region is surrounded by a pair of quarter-lateral wave reflector regions, which are designed to be antiresonant for the fundamental mode and higher order modes, which do not meet the antiresonant condition suffer large lateral losses and are suppressed. It is expected that only the fundamental mode will be supported inside ARROW VCSELs [197,198,199,200,201,202,203,204].

Figure 5-7 Schematic of an ARROW VCSEL

However, it was observed experimentally that high-order transverse LMs could still be excited in ARROW VCSELs at high injection current^[197]. This is due to the reduction of radiation loss margin (i.e., radiation loss difference between fundamental and high-order transverse LMs) by the influence of nonlinear effects such as carrier spatial hole burning (SHB)^[205] and thermal lensing^[206]. Currently, self-consistent electro-opto-thermal models have been developed to study the multi-transverse-mode operation of ARROW VCSELs^{[196], [207]}. It

was shown that the radiation losses of transverse LMs are sensitive to the effective index profile of ARROW. At high injection current, the radiation loss margin can be reduced by the nonlinear effects so that the excitation of high-order transverse LMs is unavoidable. Therefore, it is a challenge to realize much higher power single-mode lasing emission from VCSELs by using ARROW.

Hence, we decided to study the Modal characteristics of anti-resonant reflecting optical waveguide (ARROW) vertical-cavity surface-emitting lasers (VCSELs) under high current injection.

To investigate the influence of carrier SHB and thermal lensing on the radiation losses of LMs, a simple self-consistent model to study the modal characteristics of ARROW VCSELs is constructed and all the necessary nonlinear effects have been taken into consideration.

5.2.1 Laser Model

The structure of ARROW VCSELs under consideration is shown in Fig. 5.7 [197]. The center low-index core region with diameter d_1 ($= 2r_{\text{core}}$) is surrounded by two cladding layers. The first cladding layer has thickness of s and the second one has thickness of d_2 . In the conventional design of ARROW, the refractive indices of the core (n_1) and second cladding layers (n_3) are set to be the same. The refractive indices of the first (n_2) and outer cladding layers (n_4)

are also set to be the same but slightly higher than those of the core and cladding layers. In the investigation, two transverse leaky modes (i.e., LM₀₁ and LM₁₁) with the lowest radiation losses will only be considered ^[200].

The emission characteristics of ARROW VCSELs can be modeled by the following rate equations of photon density S and carrier concentration N ^[200], [207], [208]

$$\frac{dS_{mn}}{dt} = v_g [\Gamma_z \langle g_{mn} \rangle - (\alpha_{mn} + \gamma_{mn})] S_{mn} + \Gamma_z \beta_{sp} B \langle N \rangle^2 \quad (5.14)$$

$$\begin{aligned} \frac{\partial N}{\partial t} = & \frac{J}{qd} - \frac{N}{\tau_e} - v_g \sum_{mn} g |\psi_{mn}|^2 S_{mn} \\ & + D_N \left(\frac{1}{r} \frac{\partial N}{\partial r} + \frac{\partial^2 N}{\partial r^2} \right) \end{aligned} \quad (5.15)$$

where the subscripts m and n are integers standing for the transverse mode orders. ψ_{mn} is the transverse distribution of mn -order leaky mode and can be obtained from the cylindrical optical transfer matrix method ^[207]. v_g is the group velocity, Γ_z is the longitudinal optical confinement factor, α_{mn} is the total cavity loss including absorption and mirror losses, γ_{mn} is the radiation loss, β_{sp} is the spontaneous emission factor, B is the bimolecular recombination coefficient, $\langle N \rangle$ is the spatially averaged carrier concentration at time t , J is the spatially dependent injection current density, q is the electric charge, d is the thickness of the core region, and τ_e is the carrier lifetime. The last term on the right hand side of (5.15) stands for the effect of carrier diffusion, and D_N is the carrier diffusion coefficient. The modal gain $\langle g_{mn} \rangle$ is defined as

$$\langle g_{mn} \rangle = \frac{\int_0^\infty g |\psi_{mn}|^2 r dr}{\int_0^\infty |\psi_{mn}|^2 r dr}. \quad (5.16)$$

The azimuthal-dependence of ψ_{mn} has been ignored in the calculation. The optical gain of quantum well material used for the VCSELs is modeled by a logarithm form ^[209]

$$g = \alpha_N \ln(N / N_T) \quad (5.17)$$

where α_N and N_T are the temperature-dependent gain coefficient and transparent carrier concentration, respectively. α_N and N_T can be approximated by ^[210]

$$\alpha_N = a_0 + a_1 T + a_2 T^2 + a_3 T^3 \quad (5.18)$$

$$N_T = b_0 + b_1 T + b_2 T^2 \quad (5.19)$$

where $a_0, a_1, a_2, a_3, b_0, b_1$, and b_2 are constant parameters.

The spatially dependent injection current density J can be approximated by

$$J = \begin{cases} J_0 & r \leq r_{core} \\ J_0 \exp[-(r - r_{core}) / r_0] & r > r_{core} \end{cases} \quad (5.20)$$

where J_0 is the injection current density within the injection region, r and r_0 denote the transverse direction and effective diffusion length of the injection current, respectively.

The temperature-dependent carrier lifetime τ_e is given by

$$\tau_e = \frac{\tau_{e0}}{\exp[(T(t) - T_0) / T_{0ss}]} \quad (5.21)$$

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where τ_{e0} is the carrier lifetime at 300 K, T_0 is the background (substrate) temperature, and T_{0ss} is the characteristic temperature. The variation of the refractive index Δn_N induced by the change of carrier concentration ΔN is given by

$$\Delta n_N = -\alpha_H \frac{\lambda_0}{4\pi} \Gamma_z \frac{a_N}{N} \Delta N \quad (5.22)$$

where α_H is the linewidth broadening factor and λ_0 is the lasing wavelength. The variation of temperature change, ΔT , along the transverse direction can be estimated by solving the heat conduction equation using Green's function method ^{[207], [211]}

$$\Delta T = \frac{4}{HW^2} \sum_{m=1}^{\infty} \sum_{n=1}^{\infty} \frac{A_{mn}}{J_0^2(L_n W)} J_0(L_n r) \quad (5.23)$$

where H is the height of the VCSELs, $W = r_{\text{core}} + s + d_2$, J_0 is the zero-order Bessel function of the first kind, and $L_n = \eta_n/d_1$, η_n are the roots of $J_1(L_n W) = 0$. A_{mn} are a set of parameters used to calculate the temperature rise by considering the heat in all longitudinal layers, including the substrate, n-DBR (distributed Bragg reflector), active, and p-DBR layers. The form and computation of A_{mn} can

be found in Ref. ^[207]. Thermal-induced refractive index change Δn_T can then be expressed as

$$\Delta n_T = \frac{\partial n}{\partial T} \Delta T \quad (5.24)$$

where $\partial n / \partial T$ is the linear approximation to the refractive index change due to the temperature change ^[207]. Hence, the total change of refractive index, Δn , is given by $\Delta n = \Delta n_N + \Delta n_T$.

The output power of the leaky modes, P_{mn} , can be calculated by

$$P_{mn}(t) = \frac{1}{2} h \frac{C}{\lambda} v_g (1 - R_{DBR}^2) \pi a_{core}^2 S_{mn}(t) \quad (5.25)$$

where h is the Plank's constant, C is the light speed in vacuum, and R_{DBR} is the reflectivity of p-DBR. The typical device parameters of ARROW VCSELs used in this calculation are listed in Table 5-1. We use InGaAs based material systems for the calculations as the results can be compared with the experimental finding. It is believed that the design of ARROW VCSELs using other wide bandgap materials will be similar to the use of InGaAs based materials. Therefore, the presented results will be used as a reference for the design of ZnO based ARROW VCSELs in our further studies of wide bandgap microcavities.

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Parameter	Symbol	Value
lasing wavelength	λ_0	980 nm
radius of core region	r_{core}	4 μm
thickness of core region	d	0.4 μm
longitudinal optical confinement factor	Γ_z	0.06
carrier lifetime	τ_{e0}	2 ns
bimolecular recombination coefficient	B	$1 \times 10^{-10} \text{ cm}^3/\text{s}$
spontaneous emission factor	β_{sp}	1×10^{-5}
linewidth enhancement factor	α_{H}	3.7
group velocity	v_{g}	$0.833 \times 10^{10} \text{ cm/s}$
reflectivity of p-DBR	R_{DBR}	0.997
characteristic temperature	T_{0ss}	40 K
carrier diffusion coefficient	D_{N}	$10 \text{ cm}^2/\text{s}$
electric charge	q	$1.6 \times 10^{-16} \text{ C}$
current diffusion length	r_0	0.5 μm
total cavity loss of LM ₀₁	α_{01}	50 cm^{-1}
total cavity loss of LM ₁₁	α_{11}	50 cm^{-1}
thermal-induced index-change coefficient	$\partial n / \partial T$	$3 \times 10^{-4} \text{ K}^{-1}$
gain parameter	a_0	-46963.3 cm^{-1}
gain parameter	a_1	$371.56 \text{ cm}^{-1}\text{K}^{-1}$
gain parameter	a_2	$-0.941029 \text{ cm}^{-1}\text{K}^{-2}$
gain parameter	a_3	$0.99274 \times 10^{-4} \text{ cm}^{-1}\text{K}^{-3}$
parameter for transparent carrier	b_0	$2.723 \times 10^{18} \text{ cm}^{-3}$
parameter for transparent carrier	b_1	$-2.417 \times 10^{16} \text{ cm}^{-3}\text{K}^{-1}$
parameter for transparent carrier	b_2	$6.4786 \times 10^{13} \text{ cm}^{-3}\text{K}^{-2}$

Table 5-1 Device parameters used in the calculation

5.2.2 Results and discussion

A. Steady-state Characteristics of Conventional ARROW VCSELs

In conventional design of ARROW^[198,200,201], the refractive indices of the core region (n_1), first (n_2), second (n_3), and outer cladding layers (n_4) are taken to be 3.3, 3.35, 3.3, and 3.35, respectively. The dimensions of the first and second cladding layers (i.e., the values of s and d_2) are selected so that LM_{01} satisfies resonant oscillation inside the ARROW (i.e., LM_{01} experiences the lowest radiation loss). For ARROW VCSELs with $r_{\text{core}} = 4 \mu\text{m}$, the values of s and d_2 are chosen to be 1.231 and 2.725 μm , respectively. The corresponding radiation losses of LM_{01} and LM_{11} , γ_{01} and γ_{11} , are found to be 0.527 and 6.675 cm^{-1} , and the radiation loss margin $\Delta\gamma (= \gamma_{11} - \gamma_{01})$ is equal to 6.148 cm^{-1} .

Figure 5-8 shows the light-current (L-I) curves of the ARROW VCSEL operating with various substrate temperatures ($T_0 = 300, 320, 340$, and 360 K). During the calculation, the influences of carrier SHB and thermal lensing are taken into consideration. It is noted that the threshold current density J_{th} increases with the increase of T_0 and LM_{11} mode is excited at a high injection level. Further increase of J_0 saturates the total output power and the peak total power decreases with the increase of T_0 .

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Figure 5-8 L-I curves of conventional ARROW VCSEL at different T_0

In order to understand the excitation mechanism of high-order modes, Figure 5-9 (a), (b), (d), and (e) display, respectively, the profiles of ΔT , Δn_T , N , and Δn_N versus J_0 and r at $T_0 = 300$ K. Fig. 3(c) and (f) also show the variation of $\Delta n_T(r = 0)$ and $\Delta n_N(r = 0)$ versus J_0 for different values of T_0 . It is noted that the values of $|\Delta n_N|$ and Δn_T increase with the increase of J_0 . This is because carrier SHB reduces the refractive index (i.e., defocusing effect), while the thermal lensing effect increases the refractive index (i.e., focusing effect) near the core region of the laser. The value of Δn_T increases more rapidly than that of $|\Delta n_N|$ with the increase of J_0 as the influence of thermal lensing effect dominates over carrier SHB especially at high injection current.

Figure 5-9 Plots of (a) ΔT , (b) Δn_T , (d) N , and (e) Δn_N versus J_0 and r for $T_0 = 300$ K. Plots of (c) $\Delta n_T(r = 0)$ and (f) $\Delta n_N(r = 0)$ versus J_0 and T_0 .

The dependences of γ_{01} , γ_{11} , and $\Delta\gamma$ on the influence of carrier SHB and thermal lensing are shown in Figure 5-10. Fig. 5-10 (a), (b), and (c) plot the variations of γ_{01} , γ_{11} , and $\Delta\gamma$ versus J_0 for different values of T_0 . The results calculated without the consideration of nonlinear effects are also given in the figure (i.e., red horizontal lines) for comparison. It is observed that the values of γ_{01} , γ_{11} , and $\Delta\gamma$ at $J_0 = 2$ kA/cm² are much higher than those calculated without consideration of nonlinear effects (i.e., red lines). The increase of γ_{01} , γ_{11} , and $\Delta\gamma$ is due to the dominance of carrier SHB (i.e., defocusing effect) at low injection current (i.e., the region on the left hand side of the dot lines shown in Fig. 5-10). On the other hand, the values of γ_{01} , γ_{11} , and $\Delta\gamma$ decrease with the increase of J_0 . This is because thermal lensing effect (i.e., focusing effect) dominates over carrier SHB at high injection current (i.e., the region on the

right hand side of the dot lines shown in Fig. 5-10). Therefore, it can be concluded that thermal lensing effect is the dominant mechanism for the excitation of LM_{11} due to the reduction of $\Delta\gamma$ at large J_0 .

Figure 5-10 Plots of (a) γ_{01} , (b) γ_{11} , and (c) $\Delta\gamma$ versus J_0 for different values of T_0 . Red lines in (a), (b), and (c) refer to γ_{01} , γ_{11} , and $\Delta\gamma$ calculated without the consideration of nonlinear effects, respectively.

B. Searching for Optimum s and d_2 for Maximum Value of $\Delta\gamma$

It has been shown that the value of radiation loss margin $\Delta\gamma$ can be increased by carefully selecting the values of s and d_2 in the design of ARROW^[201]. This can be done by searching for a maximum value of $\Delta\gamma$ over the ranges of s and d_2 . Figure 5-11 displays the contour plot of $\Delta\gamma$ versus s and d_2 for the ARROW with conventional design given in section A. Table 5-2 lists 3 possible sets of s and d_2 for 3 local maximums of $\Delta\gamma$ marked by white asterisks in Figure 5-11. It is observed that the radiation loss of LM_{01} remains relatively low ($< 1 \text{ cm}^{-1}$) in the selection of s and d_2 . However, compared with the conventional design (i.e.,

the black asterisk in Figure 5-11), a much larger value of $\Delta\gamma$ can be obtained.

Figure 5-11 Contour plot of $\Delta\gamma$ versus s and d_2 . Three peak values of $\Delta\gamma$ are indicated in the figure using white asterisks. The value of $\Delta\gamma$ obtained from the conventional design is indicated by a black asterisk.

s (μm)	d_2 (μm)	γ_{01} (cm^{-1})	γ_{11} (cm^{-1})	$\Delta\gamma$ (cm^{-1})	J_{th} (kA/cm^2)
2.75	3.7	0.963	34.638	33.675	~ 2.58
1.90	3.7	0.913	35.957	35.044	~ 2.56
1.05	3.7	0.895	37.054	36.159	~ 2.55

Table 5-2 Possible values of s and d_2 for maximum value of $\Delta\gamma$.

From Table 5-2, it may be expected that ARROW with a large initial value of $\Delta\gamma$ can sustain stable single-mode operation in VCSELs. However, the effective index profile of ARROW as well as the corresponding $\Delta\gamma$ can be changed due to the influence of nonlinear effects (as shown in Figure 5-10). Figure 5-12 plots the corresponding L-I curves and variation of $\Delta\gamma$ with J_0 . The L-I curves and variation of $\Delta\gamma$ for the conventional ARROW VCSEL are also plotted in

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the figures (blue color lines) for comparison. It is observed that the new design of ARROW (i.e., using the parameters of s and d_2 given in Table 5-2) can slightly increase the maximum value of J_0 (by a maximum value of 2.9 kA/cm^2) for single-mode operation when compared to its conventional design; however, LM_{11} can still be excited at high injection current. This is because the thermal lensing effect significantly reduces $\Delta\gamma$ at high injection current and the rapid reduction of $\Delta\gamma$ cannot be avoided using the different choices of s and d_2 . In fact, similar results (i.e., delay by $0.5 \times J_{\text{th}}$ for the excitation of LM_{11}) have also been reported in Figure 7 of Ref. [201].

Figure 5-12 [(a), (c), and (e)] L-I curves and [(b), (d), and (f)] γ - J_0 curves of ARROW VCSELs with the design given in Table 2, where $d_2 = 3.7 \text{ }\mu\text{m}$, solid and dash lines correspond to LM_{01} and LM_{11} , respectively. Blue lines give the results of the conventional design for comparison.

It is found that the reduction of radiation loss margin at high injection current cannot be improved only by optimizing the dimensions of ARROW. Therefore, a new ARROW structure with a step index profile was proposed to sustain stable single-mode operation in VCSELs. It is verified that single-mode operation can be achieved in VCSELs at high injection current if the dimensions and refractive index profile of ARROW are selected appropriately. This design may lead to even higher single mode power (> 10 mW) in ARROW VCSELs.

C. New Design for Refractive Index Profile of ARROW

From Figure 5-9, it is noted that

- 1) the variation of $|\Delta n_N|$ is often less than 0.01 but that of Δn_T can be as large as 0.03, 2) the refractive index profile of Δn is higher at the core region than that at the cladding layer under the condition of high current injection, and
- 3) the reduction of $\Delta\gamma$ is due to the larger increase of effective refractive index inside the core region of the ARROW.

Hence, it is suggested to modify the refractive index profile of the ARROW as follows: $n_1 = 3.3$, $n_2 = 3.4$, $n_3 = 3.35$, and $n_4 = 3.4$. In this design, the refractive indices of all cladding layers are increased by 0.05 in order to compensate the change of Δn at high injection current. Furthermore, according to the design rule of conventional ARROW^[192], the values of s and d_2 are calculated (i.e., by minimizing the value of γ_{01}) to be 0.873 and 0.437 μm , respectively. The

corresponding values of $\Delta\gamma$ (γ_{01}) are found to be 12.461 (8.247) cm^{-1} . Although the value of γ_{01} is increased by more than 7.7 cm^{-1} when compared to the conventional design given in section IIIA, the value of $\Delta\gamma$ can also be increased by 6.3 cm^{-1} . It is also recorded that the corresponding threshold current density is increased by less than 0.2 kA/cm^2 compared with the results of Figure 5-12. Figure 5-13(a) plots the L-I curves of VCSELs with the new refractive index design of ARROW. The corresponding radiation losses of both modes versus J_0 are also shown in Figure 5-13(b). It is observed that stable single-mode operation is sustained in the ARROW VCSELs and the values of γ_{01} and γ_{11} as well as $\Delta\gamma$ maintain almost unchanged over the range of J_0 . However, the output power is saturated to less than 1.4 mW.

Figure 5-13 Plots of (a) L-I curves and (b) variation of γ versus J_0 of VCSEL with new design of refractive index profile of ARROW, where $J_{\text{th}} \approx 2.75$ kA/cm^2 . Solid and dash lines correspond to LM_{01} and LM_{11} , respectively. Red lines denote the values of γ_{01} and γ_{11} calculated without the consideration of nonlinear effects.

Figure 5-14 plots the variations of (a) ΔT , (b) Δn_T , and (c) Δn_N versus J_0 and r of the ARROW VCSEL. The rise of ΔT leads to a large value of Δn_T (i.e., near 0.03) at $J_0 = 16 \text{ kA/cm}^2$ and the highest value of $|\Delta n_N|$ is about 0.004 (i.e., this value is similar to that of the conventional design). Hence, the new refractive index profile of ARROW can tolerate the change of Δn_T so that the variation of $\Delta \gamma$ versus J_0 is negligible. On the other hand, the size reduction of s and d_2 increases the value of ΔT in the active region so that the maximum output power reduces to 1.4 mW. It is noted that the value of $\Delta T(r = 0)$ can be as high as 50 K at $J_0 = 10 \text{ kA/cm}^2$.

Figure 5-14 Plots of (a) ΔT , (b) Δn_T , and (c) Δn_N versus J_0 and r for ARROW with new design of refractive index profile.

D. Optimizing the dimensions of ARROW with new design of refractive index profile

In section 5.2.2.C, the problem of the reduction of $\Delta \gamma$ with the increase of J_0 has been solved, however, at the expense of low output power. This is because the small values of s and d_2 (i.e., reduction of cavity size) cause the increase of ΔT . Hence, it is necessary to obtain other large values of s and d_2 to avoid the

influence of large ΔT . Figure 5-15 gives the contour plots of (a) γ_{01} and (b) $\Delta\gamma$ in the plane of s and d_2 for the ARROW with refractive index profile given in section C. In this selection of s and d_2 , maximization of $\Delta\gamma$ is not necessary. This is because a large value of $\Delta\gamma$ is always accompanied with a large value of γ_{01} (i.e., high laser threshold). In fact, it is more important to maintain the value of $\Delta\gamma$ over a wide range of J_0 . Therefore, the key of this design is to maintain $\Delta\gamma$ (γ_{01}) at a reasonable large (small) value for some large values of s and d_2 .

Figure 5-15 Contour plots of (a) γ_{01} and (b) $\Delta\gamma$ versus s and d_2 . The values of s and d_2 chosen in types I and II optimization are given by using circle and square point, respectively.

From Figure 5-15, the value of d_2 can be selected to be $3.7 \mu\text{m}$ (i.e., to be the same as that obtained in section IIIB). To determine the value of s , there are two possible considerations as indicated in Figure 5-15. The first case, namely type I: the value of s is calculated by selecting for the second root of the first-

order Bessel function. Hence, the value of s can be expressed as

$$s = 4.611 / \left(k_0 \sqrt{n_2^2 - n_{eff}^2} \right) \quad (13)$$

where n_{eff} is the effective refractive index of ARROW. It is found that the values of s , γ_{01} , and $\Delta\gamma$ are $0.87 \mu\text{m}$, 9.17 cm^{-1} , and 15.93 cm^{-1} , respectively.

For the second case, namely type II, the value of s can be calculated from the third root of the first-order Bessel function, that is

$$s = 7.768 / \left(k_0 \sqrt{n_2^2 - n_{eff}^2} \right). \quad (14)$$

In this case, the values of s , γ_{01} , and $\Delta\gamma$ are $1.15 \mu\text{m}$, 8.32 cm^{-1} , and 14.11 cm^{-1} , respectively.

Figure 5-16 plots the L-I curves and radiation losses of the ARROW VCSELs for the cases of types I and II. It is observed that stable single-mode operation can be achieved at high injection current due to the stable value of $\Delta\gamma$ over the entire range of J_0 . In addition, the maximum output power is improved especially for the case of type II. It is worth to point out that although the maximum single-mode output power obtained in Figure 5-16 only slightly improved over the design given in Figure 5-12, single-mode operation can be obtained at much higher injection current. Hence, if the saturation power can be improved (i.e., by using heatsink to reduce the influence of thermal effects), it is possible to achieve high-power single-mode operation at high injection current.

Figure 5-16 L-I and γ - J_0 curves for ARROW with new design of refractive index profile and dimensions, where $J_{th} \approx 2.77$ kA/cm², $d_2 = 3.7$ μ m, circle lines: $s = 0.873$ μ m (type I), and square lines: $s = 1.471$ μ m (type II).

Figure 5-17 compares the variation of ΔT and Δn_T versus J_0 and r for the cases of type I, II and that obtained in section IIIC. It is noted that the design proposed in this section reduces the influence of ΔT so that the peak output power can be improved. Hence, we have shown that stable single-mode operation of VCSELs can be achieved by using ARROW.

Figure 5-17 Plots of (a) ΔT and (b) Δn_T versus J_0 and r for ARROW with new design of refractive index profile and dimensions; $d_2 = 3.7 \mu\text{m}$, type I: $s = 0.873 \mu\text{m}$, type II: $s = 1.471 \mu\text{m}$.

The proposed ARROW VCSEL can be implemented by optimizing the dimension of the etch-stop layer shown in Figure 5-7. As the refractive index of the etch-stop layer is higher than that of the DBRs, the effective refractive index of the high-index cladding layers, Dn_B , can be obtained by varying the thickness of the etch-stop layer ^[11].

In conclusion, a simple self-consistent model is developed to study the modal characteristics of ARROW VCSELs. Using the model, we have verified that:

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- The modal characteristics of ARROW VCSELs are mainly dependent on the influence of carrier SHB (thermal lensing) at the condition of low (high) injection current;
- The excitation of high-order transverse LMs in ARROW VCSELs at high injection current is due to the influence of thermal lensing;
- It is noted that the thermal induced refractive index change inside the active layer of VCSELs alters the resonant condition of ARROW so that the value of $\Delta\gamma$ is also be reduced. Therefore, stable single-mode operation of VCSELs cannot be obtained at high injection current only by optimizing s and d_2 of ARROW for a maximized value of $\Delta\gamma$.

To sustain stable single-mode operation in ARROW VCSELs under the condition of high injection current, it is proposed to modify the refractive index profile of ARROW from its conventional design. This can be done by

- Increasing the refractive indices of the cladding layers of ARROW by 0.05 in order to compensate any change of Δn_T caused by thermal lensing effect;
- Selecting larger values of s and d_2 in order to reduce the amount of heat generated inside the laser cavity. In addition, ARROW with large values of s and d_2 is easier to be fabricated than that with small values of s and d_2 .

Hence, there is no doubt that VCSELs can sustain stable single-mode operation at high injection current by using ARROW provided that the dimensions and refractive index profile of ARROW are properly selected.

It is found that the influence of carrier spatial hole burning increases the radiation loss margin of ARROW at low injection current, while the thermal lensing effect significantly reduces radiation loss margin of ARROW at high injection current. Hence, the excitation of multiple-transverse modes in VCSELs is inevitable under high injection current even if the dimensions of the ARROW have been optimized to maximum radiation loss margin.

Therefore, it is proposed to modify the refractive index profile of the ARROW to compensate against the influence of thermal lensing in VCSELs. It can be shown that the radiation loss margin of the modified ARROW can maintain unchanged under the influence of thermal lensing if the dimensions and refractive index profile of the ARROW are properly designed. Hence, stable single-mode operation can be obtained in ARROW VCSELs at high injection current.

Chapter 6

ENHANCEMENT OF RANDOM LASER WITH BRAGG REFLECTOR

6.1 Introduction

With the successful growth and the precious thickness control of ZnO and Al₂O₃ thin film deposition, we proposed to improve the Random laser emission from ZnO thin film with a disturbed Bragg reflector (DBR) in this chapter. DBR is a periodic structure, made of multiple pairs of different refractive index dielectric materials and each layer has a thickness equal to a quarter of the designed wavelength. The high reflectivity of the DBR at UV range (< 400nm) will enhance the reflection of the facet in between ZnO and the substrate. Therefore, it is expected to improve the optical confinement and as a result, the random laser performance.

As mentioned in the introduction chapter, the fabrication of UV lasers with 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 large number of random modes limited the usefulness of ZnO thin-film Random lasers for practical applications [212]. 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 [213]. 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 [214].

We propose the realization of vertical-cavity random lasers by depositing a polycrystalline ZnO thin film on to a DBR. The polycrystalline ZnO thin film, which supports random lasing action at ~ 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 1st bandedge energy on the high energy side of the reflection spectrum of the vertical-cavity Random laser. As a result, random modes radiate away from the 1st 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

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similar to that using 2D photonic crystal to control the lasing wavelength of microcavity lasers ^[215]. This is because the DBR structure is a photonic crystal in 1D.

6.2 Fabrication Procedures

Figure 6-1 (a) Schematic of the vertical-cavity Random laser. (b) The TEM image of the $\text{Al}_2\text{O}_3/\text{ZnO}$ DBR fabricated by the FCVA technique.

Figure 6-1(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^[216]. 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

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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. Figure 6-1(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 figure 6-2. 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.

Figure 6-2 Normal reflection spectrum of the DBR – comparison between measurement and calculation.

There are 3 reasons to fabricate the DBR using ZnO and Al_2O_3 as the dielectric materials:

1) The contrast of refractive 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 ^[217]. Hence, the use of Al₂O₃/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 FCVA technique at room temperature do not support random lasing action ^[216]. 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 1st bandedge energy matches with the peak gain wavelength of the polycrystalline ZnO thin film. 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 ^[213]. 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.

6.3 Results and Discussions

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. Figure 6.3 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/cm² (3.3 MW/cm²) 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.

Figure 6-3. 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.

Figure 6-4a shows the emission spectra measured from the surface of the film without DBR at room temperature. For pump intensities exceeding 5.1 MW/cm², sharp peaks (i.e., random modes) are excited at around 385 nm. The mechanism of radiative recombination inside the polycrystalline ZnO thin film may be attributed to the process of exciton-exciton scattering^[217]. 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^[213]. For pump intensities at about 2.9 times its threshold, 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 ^[218].

Figure 6-4 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 figure 4(b). The Label (A, B and C) refers to the emission spectrum captured at the excitation power with reference to figure 6-3

Figure 6-4b shows the emission spectra of the film with DBR with spectrum A, B and C with respect to below threshold, at threshold and above threshold stated at figure 6-3. The reflection spectrum of the polycrystalline ZnO thin

film on the DBR is also plotted in the figure. It is observed that the 1st bandedge energy of the reflection spectrum is located at 385 nm. A small dip is also appeared 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 1st bandedge energy. This is due the peak gain wavelength of the polycrystalline ZnO film matches with the 1st 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.

Figure 6-5 plots (see figure on the right hand side) the calculated electric field intensity profile inside the vertical-cavity Random lasers with wavelength λ

equal to ~ 385 nm.

As shown in figure 6-5, 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. A standing wave has established within the structure and the DBR has enhanced the confinement of light with the cavity, 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 1st bandedge spectrum.

6.4 Summary

In this chapter, a simple one-step fabrication process using FCVA technique was proposed to fabricate a polycrystalline ZnO thin film on an $\text{Al}_2\text{O}_3/\text{ZnO}$ 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 exhibits 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.

Chapter 7

ZnO/ZnMgO MULTIPLE QUANTUM WELL RIDGE WAVEGUIDE LASERS

7.1 Introduction

Upon the successful fabrication of DBR structure, we decided to improve our random laser device further by incorporating with multiple quantum wells structure. Although high quality ZnO based QW semiconductor lasers have been fabricated by pulsed laser deposition ^[219,220] and molecular beam epitaxy ^[221,222] techniques, the stringent requirement of lattices matching substrate and high substrate temperature ($> 500\text{ }^{\circ}\text{C}$) have refrained the mass production of ZnO QW lasers at low cost. Therefore, FCVA technique is proposed to fabricate ZnO based QWs semiconductor lasers. The advantage of this technique is that it allows the deposition of ZnO/ZnMgO QWs on quartz substrate at substrate temperature of 200°C . Our objective is to fabricate a ZnO QW lasers at low temperature but yet, its room temperature optical

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characteristics of ZnO QW lasers can be comparable with those obtained from the mainstream fabrication methods ^[221,222].

QW laser is analogue with conventional semiconductor laser, except for the fact that the thickness of the active region is comparable to the de Broglie wavelength of the carriers and leads to quantum confinement. This confinement alters the density of the states (DOS) from bulk semiconductor (i.e. increases as the square root of the energy) to a 2 dimensions (i.e. step-like functions). As a result, population inversion for QW laser can be easily achieved and therefore, it has a better gain per injected carrier than conventional lasers and results in lower thresholds current ^[223]. In addition, QW lasers provide higher differentiate gain, reduced laser linewidth and superior mode stability ^[224].

Due to the complexity of the theoretical study, the design and modeling were calculated by another member from our group. For the completeness of this thesis, part of his works is cited here ^[225]. In his work, the energy bands of the ZnO / ZnMgO QWs are modeled with the assumption that the conduction and valence bands are decoupled, by using the *k.p* method with a 6 x 6 Hamiltonian to account for the spin-orbit coupling which result in the degenerate A-, B- and C-hole bands ^[226]. The free exciton optical gain and the momentum matrix ^[227] are deduced and hence, the optimums design of the QW structure with maximum peak gain, as a result of the maximum oscillator strength ^[228]. Figure 7-1 ^[229] shows the plot of oscillator strength with various well widths. The optimum well width, L_w , is found to be 1.5nm for a wide range of Mg mole

fraction. The present of this optimum well width is attributed on the electronic confinement of excitons, which is dependent on the amount of overlapping between the electron and hole wave function.

Figure 7-1. Total oscillator strength of ZnO-Mg_xZn_{1-x}O QWs for x= 0.12 and 0.27 as a function of well width.

Unfortunately, our experiment setup is not sensitive enough for the detection of radiation emission from Single Quantum Well (SQW) laser. Therefore, our focus will be on Multiple Quantum Well (MQW) lasers, in the hope of stronger emission intensity^[230]. Apart from its emission intensity, when compared with SQW lasers, MQW lasers provide a lower carrier density when maximum optical confinement is available. As a good approximation, a separate confinement MQW laser has a linear relationship between the waveguide confinement factor and the number of quantum wells^[224]. Ideally, the lowest

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threshold current for MQW laser is achievable at the highest QW number that can be accommodated in the optical waveguide, with the cavity length adjusted to the corresponding shortest optimum value ^[223]. However, MQW lasers are more strongly degraded by interface non-radiative recombination at crystal defects, mainly at the well-barrier interfaces and the fluctuation of the layer thickness.

For our experimental study, ZnO/ZnMgO multiple quantum wells (MQWs) thin-film waveguides with ridge structures have been fabricated on quartz substrates. Low-temperature deposition of high-quality ZnO/ZnMgO MQWs thin films was achieved by filtered cathodic vacuum arc technique. A ridge is defined on the thin film by plasma etching. Room temperature lasing with peak wavelength at 378 nm of 1.5 nm well width was observed under 355 nm optical excitation. Exciton-exciton scattering was attributed to the amplified spontaneous emission observed from the MQWs waveguide. The net optical gain can be larger than 80 cm^{-1} at a pump intensity of 2 MW/cm^2 .

7.2 Fabrication Procedures

Figure 7-2 Schematic of the ZnO/ZnMgO MQWs and the calculated intensity profile of the confined light along the growth direction of the MQWs. The inset shows the TEM image of the ZnO/ZnMgO MQWs.

Figure 7-2 shows the schematic diagram of a ZnO /ZnMgO multiple quantum wells (MQWs) structure. The MQW consists of 10 ZnO wells with thickness varies from 1.5nm to 5nm and the ZnMgO barrier has thickness of 10 nm. ZnO and ZnMgO were deposited separately on a quartz substrate from two independent cathodic arc sources from a modified FCVA system ^[231]. Zn and Zn:Mg (with 15 at% of Mg) metal targets, both with purity of 99.99%, were used to form Zn and Zn:Mg plasmas respectively from the two cathodic arc sources. Oxygen gas was introduced from an outlet which was 2 cm above the surface of the substrate. The gas was mixed with the metal plasmas to form metal-oxide thin films. The thickness of the metal-oxide thin films can be

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controlled by a gold crystal thickness monitor. In the 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 Zn:Mg cathodic arc sources were both kept at 70 A. The substrate was set to 200°C with a rotational speed of 30 rev/min. The deposition rate of the ZnO and ZnMgO were found to be 1.0 nm/min respectively.

The MQW was designed and fabricated to confine light along the growth direction of the QWs region. A 50 nm $\text{Zn}_{0.85}\text{Mg}_{0.15}\text{O}$ buffer layer was first grown on the quartz substrate and followed by 10 periods of ZnO/ZnMgO MQWs. A capped layer of ZnMgO was then deposited on the top of MQWs to provide a further confinement of light. The calculated intensity profile of the confined light along the growth direction of the MQWs is also plotted in figure 7-2. The inset of figure 7-2 shows the transmission electron microscope (TEM) image of the ZnO/ZnMgO MQWs with well width of 5 nm. As the concentration of Mg is only 15%, the MQWs structure is barely observed from the TEM image. Lattice constant along c axis of ZnO well and ZnMgO barrier were found to be 0.517 and 0.52 nm respectively.

7.3 Results and Discussions

Figure 7-3 Emission spectra of the ZnO/ZnMgO MQWs with various well widths. The inset shows the calculated and measured emission energies of ZnO/ZnMgO MQWs versus well widths.

The room-temperature optical characteristics of the ZnO/ZnMgO MQWs 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 cylindrical lens to focus a $1 \times 0.5 \text{ mm}^2$ pump stripe on the surface of the MQWs. Figure 7-3 shows the normalized emission spectra measured from the edge of the MQWs with different well width. The corresponding excitation power was maintained at $\sim 0.4 \text{ MW/cm}^2$. The

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normalized emission spectra of ZnO and ZnMgO thin films were also plotted in the figure for comparison. As expected, the emission peak is blue shifting with the reduction of well width. The inset of figure 7-2 plots the measured emission energy versus well width of the MQWs. The calculated free exciton recombination energy versus well width of the MQWs ^[232] is also plotted in the inset of figure 7-3. It is observed that the two curves shown the same trend except they are different by ~0.15 eV. This indicated that the recombination mechanism of the MQWs is mainly due to exciton-exciton scattering (EES) recombination ^[233].

In order to improve the optical characteristics of the ZnO/ZnMgO MQWs, a ridge waveguide structure is fabricated. The ridge design is used because:

- 1) It is simple to be implemented by lithographic techniques,
- 2) It improves the confinement of light in the lateral direction,
- 3) It can be easily integrated with other optical and electrical components, and
- 4) Its geometry is similar to that of the excitation pump strip.

Since the height of the waveguide has already been determined earlier, the main consideration for the ridge waveguide structure will be the etching depth and the width of the waveguide. Due to the limitation of our lithography techniques (minimum resolution of ~1 μm), the design of the waveguide structure will be focus on 1) the lateral confinement coefficient Γ_{cy} or the overlapping area between the signal and pump intensities inside the structure to be maximum ^[234]. 2) Minimum amount of lateral modes supported ^[235]. Figure 7-4 shows the

calculated variation of Γ_{cy} versus the width of the ridge waveguide with only the fundamental mode at the center of the emission spectrum (380nm) is considered. Since the number of supporting modes is inversely proportional with the cross-section area of the waveguide, we have chosen 2 μm as the design of the ridge structure as width that is narrower than the suggested value suffers largely in the confinement factor.

Figure 7-4. Lateral confinement coefficient Γ_{cy} versus width of the ridge structure

To fabricate the ridge waveguide structure, photoresist stripes with width of 2 μm were formed on the MQWs by photolithography technique. The unmasked region of the MQW is etched away by the Ar^+ ion beam sputtering system at a rate of 10nm/min. Figure 7-5 shows the light-light curves of the ZnO/ZnMgO MQWs ridge waveguide with 1.5 nm well width for both TE and TM modes. The light-light curves from the MQWs thin films are also plotted for

comparison. It is observed that the TE mode is stronger than the TM modes from both samples. This is expected as the ridge and planar waveguides are both favored TE modes. Furthermore, the confinement of TE mode is improved with the ridge structure so that the emission intensity of ridge waveguide is higher than that of planar waveguide. In fact, the slope efficiency of TE modes of the ridge guided structure is double to that of the planar structure. Kinks were also observed at about 0.5 MW/cm² (0.4 MW/cm²) for the sample without (with) ridge waveguide structure.

Figure 7-5 Light-light curves of the ZnO/ZnMgO MQWs with and without ridge waveguide 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 ZnO/ZnMgO MQWs.

Figures 7-6a and 7-6b show the emission spectra from the MQWs without and

with the ridge waveguide structure, respectively. In figure 7-6b, it is noticed that when the pump intensities exceed threshold, narrowing of emission spectra is observed. Together with the present of the kink in the light-light curves earlier, it is suggested that the lasing process is due to amplified spontaneous emission.

Figure 7-6 Emission spectra of the ZnO/ZnMgO MQWs with and without ridge waveguide structure. Label (A,B and C) refers to the spectrum measured at the excitation with reference to figure 7-5.

The cause of the emission improvement can be explained in Figure 7-7, a comparison of the measured optical gain of MQWs with and without the waveguide structure. The net optical gain was measured by variable stripe length method ^[236]. It is observed that both TE and TM net optical gains of the MQWs with ridge waveguide structure are almost 2 times larger than that without the ridge waveguide structure; as such, the MQW with ridge waveguide structure will has a stronger optical amplification and results in a lower threshold value ^[237]. The maximum gain of the TE emission from the Ridge waveguide was measured to be about 80 cm⁻¹ at 2 MW/cm².

Figure 7-7 Net optical gain of the ZnO/ZnMgO Multiple Quantum wells thin films with and without DBR structure.

7.4 Conclusion

In conclusion, we have demonstrated the deposition of high-quality ZnO/ZnMgO MQWs thin films on quartz substrates by the FCVA technique at 200°C. High intensity ultraviolet lasing is observed from the devices at room temperature due to amplified spontaneous emission. Furthermore, a ridge waveguide structure is proposed to improve the lasing performance of the MQWs. It was shown that the ridge waveguide structure can maintain stable TE mode emission at high pump intensity and the corresponding lasing threshold (slope efficiency) can be reduced (increased) by 1.25 times (a doubled). The corresponding net optical gain can be as high as 80 cm^{-1} at a pump intensity of 2 MW/cm^2 .

Chapter 8

Conclusions and Recommendation

8.1 Conclusions

Upon the completion of this project, a twin arms FCVA system is developed for the fabrication of our multilayer devices. By fine-tuning its various parameters and the redirection of the magnetic field, our system is optimized to produce high-quality samples at low depositing substrate temperature. A thickness control technique has also been introduced into our system with an accuracy of 1 nm. Furthermore, together with the capability of fabricating complex structure on a lattice mismatch substrate, integration with the current Si technology is feasible.

To characterize our devices, a low temperature photoluminescence setup has built. This setup consists of a Nd:YAG laser for sample excitation at the wavelength of 266 and 355nm, depends on the various combinations of the

harmonic crystals. Our setup also allows the measurement of the low temperature sample radiation emission, which can be lower to 77 K, with liquid nitrogen as our heat exchange agent. In this setup, we have observed the low temperature phenomenon of the ZnO random laser emission such as the blue-shifting of the emission wavelength, the narrowing of the emission FWHM, the reduction in laser threshold and the transformation of the emission mechanism.

Due to the complexity of the multilayer structure, it is important to perform a simulation studies so as to provide a better understanding and control for our devices. A theoretical model is developed to study the optical behavior of Bragg reflectors, which is based on the transfer matrix method and the consideration of the limitations of our deposition technique and possible experimental errors. Although there is a presence of lattice mismatch between adjacent layers, it is still practicable to fabricate high quality DBR with high with a reduced number of dielectric pairs due to the large refractive index contrast. Also, its optical reflectivity is comparable with other fabrication techniques. Meanwhile, the model is revised and employed for the studies of the ARROW VCSELs. With this model, we have verified that the excitation of the high order transverse LMs at high injection current is mainly due to the present of thermal lensing and it is unavoidable under high injection current even if the dimensions of the ARROW have been optimized to maximum radiation loss margin. As such, we have proposed to modify the refractive index profile to sustain a stable, single-mode operation ARROW VCSELs. However, the realization of this type of lasers with wide bandgap materials is still in

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progress due to the complexity in its laser structure.

To improve the performance of the ZnO random lasers, DBR has proposed to be incorporated with the ZnO polycrystalline thin film. A simple one-step fabrication process with FCVA technique has proposed to fabricate a polycrystalline ZnO thin film on a $\text{Al}_2\text{O}_3/\text{ZnO}$ multilayer DBR. The reflection spectrum of the DBR is designed to match the gain spectrum of the ZnO. The lasing behavior from the surface of the polycrystalline ZnO is studied with our PL system and compared with a sample without the optical support of the DBR. It has 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. This provides a practical low cost solution for narrow emission UV random lasers.

Experimental realization of multiple quantum wells (MQW) structure for low threshold ZnO emission is also demonstrated with our twin arms FCVA system. Even though the device is fabricated under low substrate temperature condition at 200°C, strong UV lasing is observed at room temperature as a result of amplified spontaneous emission. In addition, ridge waveguide structure is engineered to improve its performance by lowering its threshold power. This accomplishment has created a new possibility to integrate ZnO low threshold lasers with various low melting point and lattice-mismatched substrates.

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In conclusion, we have demonstrated that it is achievable to develop high-quality, complex multilayer devices with the FCVA depositing techniques. Also, with the integration of the multilayer structure, the performance of random lasers is indeed comparable with the conventional Fabry-Perot laser. Therefore, it is feasible to produce low cost commercial UV laser by incorporating multilayer structure into ZnO medium with the assisting of the FCVA system.

8.2 Recommendation

Beyond the alternatives for UV lasers mentioned in this thesis, Polariton microcavities (MC) laser will be another attractive possibility due to its near threshold-less laser operation which relies on cavity polaritons that genesis lies in the interaction between photons and excitons. Based on Bose Einstein condensation (BEC), when Polariton laser operates at its strong coupling regime, it can produce ultra-coherent light. However, Polariton has a relatively short lifetime, even with the presence of MC, it still remains as one of the main challenges for the experimental realization of such a state-of-the-art laser. The first demonstration of polariton parametric amplifier from CdTe-based MC was at a low temperature of 220 K ^[238]. Still, for room temperature Polariton device, it is necessary for exciton to survive at such a temperature before splitting itself into electron hole plasma. As such, by having a large exciton binding energy of ~60meV, ZnO becomes one of the promising candidates in realizing this threshold-less laser. Unfortunately, the fabrication of such a wide bandgap semiconductors complex laser remains as a nontrivial technology problem ^[239,240].

Recently, M. Kaliteevski *et al.* ^[241] have proposed an innovative method in providing the optical confinement of the Polariton, a possible alternative to the conventional MC. Instead of deploying two DBRs to create a MC, they have suggested to produce a confined electromagnetic mode at the boundary between a dielectric Bragg mirror and an isotropic medium with a negative dielectric constant, such as metal below plasma frequency. This particular mode is named

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as the Optical Tamm Plasmon Polariton (TPP) state and it is the result of the confinement created by the negative dielectric constant of the metal layer with the photonic stopband of the Bragg Reflectors.

As mentioned in the article, the condition of the eignemode to sustain a TPP at the interface between a metal layer and a DBR is given as,

$$r_{DBR}r_m = 1 \quad (8.1)$$

Where r_{DBR} is the amplitude reflection coefficient of the wave incident from the medium onto the DBR and r_m is the amplitude reflection coefficient of the wave incident from the medium onto the metal layer.

The reflection coefficient r_{DBR} can be calculated with the transfer matrix method by accounting the refractive indexes of the individual layers, thickness, as well as the optical absorption from the dielectric materials in the DBR. As for the reflection coefficient of the metal layer, r_m , Fresnel equation states that;

$$r_m = \frac{(n_a - n_m)}{(n_a + n_m)} \quad (8.2)$$

And with reference to Drude model;

$$n_m^2 = \epsilon_b \left(1 - \frac{\omega_p^2}{\omega(\omega + i\gamma)}\right) \quad (8.3)$$

Where ω is the frequency, ϵ_b is the background dielectric constant, ω_p is the plasma frequency and γ is the plasma collision rate. A more comprehensive theoretical discussion can be found in the article.

To examine the possibility in realizing such structure with our FCVA system,

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we have fabricated a simple TPP structure and its schematic is shown in Figure 8-1(a). This sample consists of a $13\frac{1}{2}$ ZnO/Al₂O₃ DBR, deposited on an UV graded quartz substrate. The top ZnO layer of the DBR is covered with a layer of Ag. For simplicity and also, the limited choices of metal with plasma frequency for UV region, we have designed our structure to resonance at visible wavelength of 510nm. Transmission electron microscopy (TEM) image of the fabricated DBR is shown in Figure 8-1(b). Again, It is observed that high optical quality ZnO/Al₂O₃ DBR can be obtained from the FCVA system at room temperature.

Figure 8-1 (a) the schematic diagram of the proposed TPP structure. (b) TEM image of the proposed fabricated structure.

Figure 8-2 compares the (a) measured and (b) calculated room-temperature reflection spectra (dashed-dotted lines) of the $13\frac{1}{2}$ pairs of DBR without Ag coating at normal incidence of light. It is observed that there are some discrepancies between the measured and calculated results. These may be due

to some fluctuation of thickness in the dielectric layers during fabrication. Nevertheless, the fabricated DBR can obtain 99% of reflectivity at ~510 nm and a bandwidth of ~60 nm for the stopband. The present of a dip in the spectral response is sufficient to demonstrate the formation of TPP at the interface between Ag and DBR by our twin arms FCVA system.

Figure 8-2 (a) Experimental result of the reflection spectra measured from the DBR with and with Ag coating. (b) Calculated result of the reflection spectra.

In conclusion, it is possible to realize UV Polariton lasers with our FCVA system by incorporating a gain medium within the TPP structure. Apart from having a low operation threshold, the complexity of the TPP lasers structure is almost reduced by half as compared with a typical MC Polariton structure. The

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remaining challenge will be to search for a negative refractive index material that is suitable for UV region and to improve the quality of the QW in the gain medium such that strong coupling can occur.

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