Journal of Crystal Growth 101 (1990) 944–952
North-Holland

BLUE SPONTANEOUS AND STIMULATED EMISSION IN VPE ZnSe EPILAYERS

X.W. FAN, Z.K. TANG and H. TIAN

Changchun Institute of Physics, Academia Sinica, Changchun 130021, People's Rep. of China

ZnSe epilayers have been heteroepitaxially deposited on GaAs substrates by vapor phase epitaxy (VPE) using hydrogen in an open tube reactor. The photoluminescence (PL) spectra and electrical properties of the ZnSe epilayers depend on the substrate temperature, Zn vapor treatment and purity of source materials. The origin of the blue emission at room temperature (RT) of the ZnSe epilayers is ascribed to exciton-carrier interaction. The pure blue electroluminescence (EL) at room temperature from a forward biased MIS diode of VPE ZnSe epilayer is reported for the first time. The blue stimulated emission, which is ascribed to exciton–exciton interactions at 77 K from a VPE ZnSe epilayer under 337.1 nm excitation from a N_2 laser, is also reported for the first time.

1. Introduction

ZnSe is a wide-gap II–VI compound which is eminently suitable material for the use in optoelectronic devices, covering the visible to the ultraviolet spectral region. In recent years, there has been considerable interest in various laboratories, including our own, in blue EL [1–3] and blue stimulated emission [4–7] in ZnSe. For such applications, the growth of high-quality epitaxial thin layers is very important.

So far, many papers have reported on epitaxial thin layers of ZnSe grown by MBE [8], MOCVD [9], LPE [10] and VPE [11]. High-quality ZnSe epilayers were grown by various methods. Although there were several reports of blue EL at RT in ZnSe MIS diodes using ZnSe epilayers grown by MBE [12,13] and on blue stimulated emission using ZnSe multilayer structures [5–7], there has been no report of blue EL and blue stimulated emission using VPE ZnSe epilayers.

In our earlier work, attention was directed to the identification of free exciton emission, to the determination of the origin of the blue EL band at RT [1,2] and to the improvement of spatial distribution of blue EL [14] in ZnSe MIS diodes operated in forward bias. Recently, effort has been devoted to the effect of purity of the ZnSe starting material and the perfection of the ZnSe crystal both on its blue EL [15,16] and on the near band edge emission originating from exciton–exciton and exciton–electron interaction in ZnSe and ZnS_xSe_{1-x} MIS diodes with high pulse current density [17,18]. In the past, our study on blue EL in ZnSe focused on bulk crystals grown by sublimation at high temperatures. The as-grown ZnSe crystals usually contained a large number of intrinsic defects, and had high resistivity even if the donor impurities had been doped. More recently, we prepared high-quality ZnSe epilayers by VPE [19–21] and MOCVD [22], and studied their optical and electrical properties.

In the present paper, the dependence of PL spectra and electrical properties of VPE ZnSe epilayers on growth conditions is discussed and the origin of the blue emission at RT of ZnSe epilayers is studied. The blue EL at RT and the blue stimulated emission at 77 K in VPE ZnSe epilayers are reported for the first time.

2. Experimental

The VPE system [19] was designed to have a large temperature gradient between the vaporization and deposition zones, and small temperature gradients in every other zone. The reactor and the furnace tubes were fabricated from quartz. High-purity ZnSe powder was used as the source material and high-purity hydrogen as the carrier

0022-0248/90/$03.50 © 1990 – Elsevier Science Publishers B.V. (North-Holland)

gas. The growth conditions of ZnSe epilayers include a substrate temperature of 300–700°C, a ZnSe source temperature of 820–830°C, a hydrogen transport flow of 40 cm^3/min and growth rates of 0.2–10 μm/h.

GaAs served as the substrate in this investigation because of its small lattice constant mismatch of 0.27% to ZnSe, as well as comparable thermal expansion coefficients. The orientation of the substrate used was (100). Before deposition the mechanically polished (100) GaAs substrates were chemically etched in a solution of $3H_2SO_3 : H_2O : H_2O_2$ at 40°C for 30 s. After etching, the GaAs substrates were thoroughly rinsed in de-ionized water. The substrates and source materials were then heated in hydrogen at low temperature for several hours.

The spectral distribution of luminescence was measured using a grating monochromator, Model SPEX 1404, with an RCA-C31034 cooled photomultiplier. The PL was excited either by the 365.0 nm line from a 250 W high-pressure Hg lamp or by the 337.1 nm line from a N_2 laser, Model UV-24, with a peak power of 3 MW/cm^2. The sample was mounted on a copper holder and immersed in a pumped liquid nitrogen cryostat with transparent windows. In this case, the temperature of the sample could be reduced down to 64 K.

3. Dependence of PL on growth conditions

3.1. Dependence of PL on substrate temperature

Fig. 1 shows the dependence of the PL spectra on substrate temperatures for as-grown ZnSe epilayers excited by the 365.0 nm line of a Hg lamp at 77 K. As seen from the figure, the edge emission region consists of two bands: E_s at 445.0 nm and FB at 460.0 nm. The former is attributed to the recombination of free excitons after interaction with free electrons in the conduction band, while the latter is associated with free-to-bound recombination. When the substrate temperature is above 600°C, the edge emission region is dominated by the FB band. On decreasing the substrate temperature, the E_s band becomes more intense and the FB band decreases. When the substrate temperature is 300°C, the edge emission region is dominated by the E_s band. This shows that the lower substrate temperature favors the E_s band, which leads to the emission band at RT [1]. The contamination from the furnace and the self-doping by Ga from the GaAs substrates are suppressed due to reducing the substrate temperature. This results in the improvement of epilayer quality and an increase of free exciton emission.

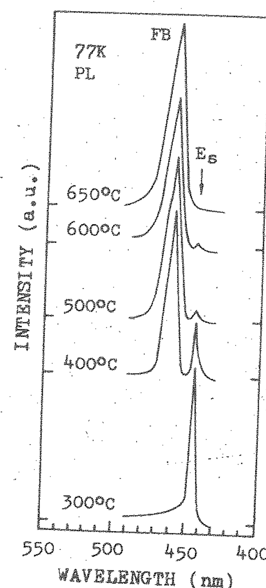


Fig. 1. PL spectra at 77 K at various substrate temperatures for as-grown ZnSe epilayers, excited by the 365.0 nm line of a 250 W Hg lamp.

3.2. Dependence of PL on Zn vapor treatment

Fig. 2 shows the dependence of the PL spectra on Zn-treatment time for ZnSe epilayers excited by the 365.0 nm line of a Hg lamp at 77 K. The PL spectrum of the as-grown ZnSe epilayers consists of the E_s band at 445.0 nm and the FB band at 460.0 nm in the edge emission region, and the D_1 band at 540.0 nm and the D_2 band at 630.0 nm in the deep center emission region. The D_2 band is attributed to a self-activated (SA) center including a Zn vacancy [19], and the D_1 band, which usually appears at high growth temperatures, may be asso-

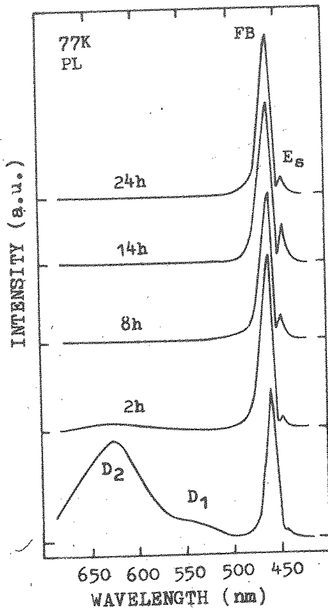


Fig. 2. PL spectra at 77 K for various Zn-treatment times for ZnSe epilayers excited by the 365.0 nm line of a 250 W Hg lamp.

ciated with contamination by some impurities [11]. As the Zn-treatment time is increased to 14 h, the E_s band becomes intense and the deep center emission bands become very weak, but as the Zn-treatment time is further increased to 24 h, the E_s band decreases. The cause of the former is considered to be that the increase of the Zn-treatment time decreases the Zn vacancy concentration, and the quality of the ZnSe epilayer improves. The latter is not understood.

3.3. Dependence of PL and electrical properties on purity of source materials

The following source materials have been used: (A) ZnSe particles of electronic grade; (B) ZnSe powder (purity analysis). Sources A and B were refined by means of sublimation in an evacuated, open chamber for two cycles. The refined sources were expressed as A_1, A_2 and B_1, B_2 for each cycle, respectively. The sources A_2 and B_2 were purified by sublimation in an evacuated and closed chamber for two (A) and three (B) cycles. The refined sources were expressed as A_3, A_4 and B_3, B_4, B_5 for each cycle, respectively. In the following, 1 to 5 serve as the number of purification cycles.

Fig. 3 shows the PL spectra of ZnSe epilayers grown with sources A to A_4. The spectrum from a ZnSe epilayer with source A shows a dominant FB band, a weak E_s band and a deep center emission band. When the sources are purified, the deep center band disappears and the E_s band becomes intense. Fig. 4 shows the PL spectra of ZnSe epilayers grown by sources B to B_5. The spectrum with B shows only a strong deep center band and no edge emission is detected. With an increase of the purification cycles, the E_s band becomes intense and the deep center band rapidly reduces.

Fig. 5 shows the dependence of the carrier concentration n, electron mobility μ and resistivity ρ of the ZnSe epilayers on the number of purification cycles of sources B. As seen from the figure, μ and ρ increase and n decreases with increasing number of purification cycles.

Muranoi and Furukoshi [23] have indicated that the conductivity of ZnSe epilayers was attributed to Ga outdiffusion from the GaAs substrate. Yao et al. [24,25] have suggested that the

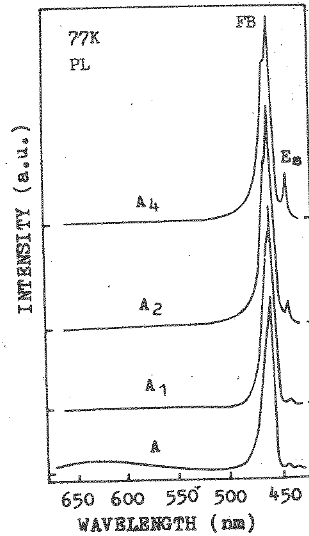
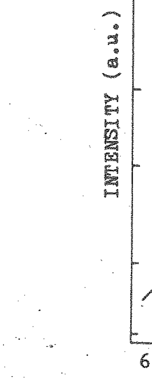
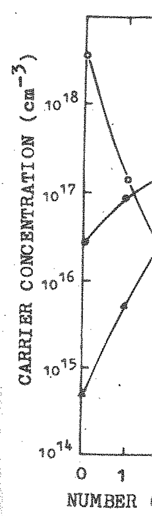


Fig. 3. Dependence of the PL spectra at 77 K from ZnSe epilayers on purity of A series source materials, excited by the 365.0 nm line of a 250 W Hg lamp.

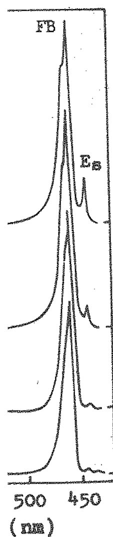




as A_3, A_4 and B_3,
vely. In the follow-
ıber of purification

a of ZnSe epilayers
'he spectrum from a
ıows a dominant FB
leep center emission
purified, the deep
ıe E_s band becomes
L spectra of ZnSe
to B_5. The spectrum
eep center band and
With an increase of
$_s$ band becomes in-
d rapidly reduces.
ence of the carrier
ility μ and resistivity
number of purifica-
en from the figure, μ
ases with increasing

[23] have indicated
e epilayers was attri-
rom the GaAs sub-
e suggested that the



ectra at 77 K from ZnSe
ce materials, excited by the
) W Hg lamp.

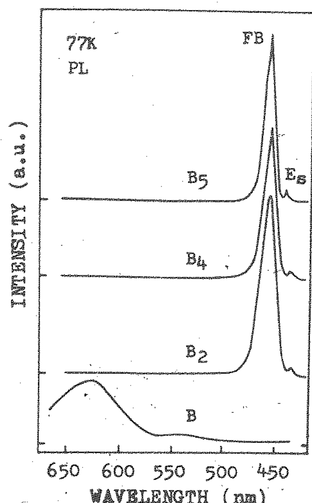


Fig. 4. Dependence of the PL spectra at 77 K from ZnSe epilayers on purity of B series source materials, excited by the 365.0 nm line of a 250 W Hg lamp.

conductivity of ZnSe epilayers mainly depended on Se vacancy native donors. Fujita et al. [26] and Yoneda et al. [27] have reported that the conductivity of ZnSe epilayers was ascribed to extrinsic donor impurities. From the results mentioned above, we suggest that the conductivity in nor-

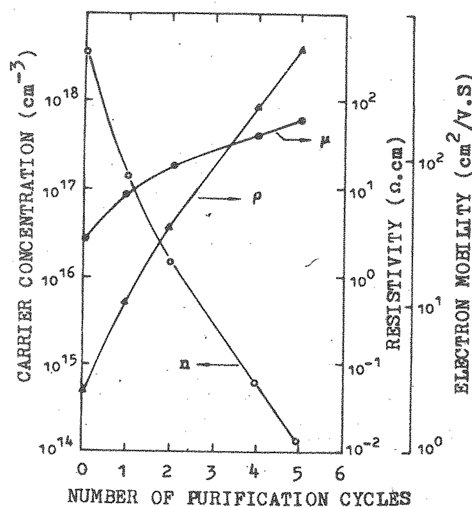


Fig. 5. Dependence of carrier concentration n, resistivity ρ and electron mobility μ at RT from ZnSe epilayers grown with B source materials on the number of purification cycles.

mally undoped VPE ZnSe epilayers is attributed to extrinsic donor impurities which mainly originate from the contamination of the ZnSe source materials, rather than from Ga outdiffusing from the GaAs substrate or from native donors. We believe that doping by appropriate shallow acceptor impurities in the high-purity and high-resistivity ZnSe epilayers should result in p-type layers.

4. PL under high excitation density

4.1. Interaction between excitons

Fig. 6 shows the PL spectra at 77 K, at various excitation densities, for ZnSe epilayers excited by the 337.1 nm line of a N_2 laser. With increasing excitation density, the E_s band at 445.0 nm broadens and its peak position shifts to lower energy. However, the FB band at 460.0 nm shows no change. It is clear that the intensity ratio R of the E_s band to the FB band depends on the excitation

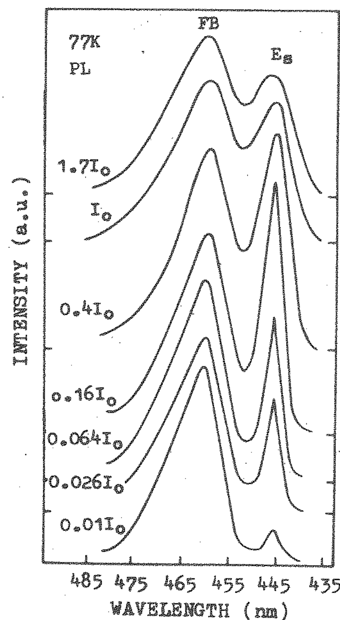


Fig. 6. Edge emission spectra at 77 K from ZnSe epilayers under various excitation densities, excited by the 337.1 nm line of a N_2 laser with $I_0 = 3$ MW/cm^2.

density. With increasing excitation density, R can be divided into two ranges: first R increases in range I and then R decreases in range II. The increase of R in range I can be explained by exciton–carrier interaction [28], but it fails to explain the result in range II. We consider that the decrease of R in range II results from a collision process of two is excitons in which one exciton is excited into the dissociation state, resulting in a free electron–hole pair, while the other recombines and produces the emission. Thus the free carrier density increases rapidly and the free exciton density increases slowly because of the dissociation of excitons under high excitation. It results in a decrease of the intensity ratio of the E_s band to the FB band.

This suggestion can be supported by the following fact. Increasing the excitation density to I_0 splits the E_s band into two bands E and P at 64 K. The position of the P band is about 20 meV below the E band; it is just equal to the binding energy of the free exciton. After two excitons collide, one of them gets an energy equal to the binding energy and becomes a free electron–hole pair. However, the other one loses the same energy and becomes a photon emission to form the P band. On the other hand, the E band is due to free exciton emission scattered by carriers. As a result, the position of the P band is the binding energy below the E band. The appearance of the P band shows clearly that the interaction between excitons occurs. It was noticed that the appearance of the P band was coincident with the beginning of range II. It follows that the interaction between excitons results in a decrease of intensity ratio of E_s band to FB band.

4.2. Nature of blue emission at RT

Fig. 7 shows near band edge emission spectra in the temperature range of 64–360 K for ZnSe epilayers excited by the 337.1 nm line of a N_2 laser. In fig. 7, the spectrum at 64 K contains two emission bands: the E band due to exciton–carrier interactions and the P band caused by exciton–exciton interaction. With increasing temperature to 77 K, only one E_s band, overlapping between the E and P bands, is detected. With a further increase

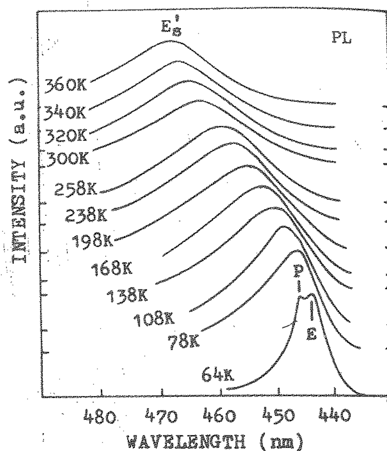


Fig. 7. Temperature dependence of the near band edge emission spectra from an as-grown ZnSe epilayer, excited by the 337.1 nm line of a N_2 laser.

in temperature, the structureless E_s band widens and shifts towards longer wavelength and finally forms the blue emission band E_s' at RT. This result suggests that the E_s' band is related to free exciton emission.

Fig. 8 shows the dependence of the E_s band intensity of the ZnSe epilayers on temperature. It

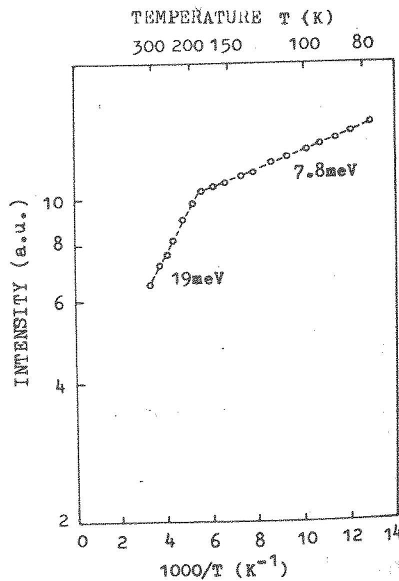


Fig. 8. Temperature dependence of the E_s band intensity from the ZnSe epilayer.

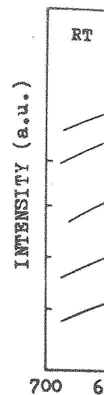
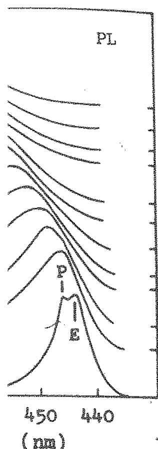


Fig. 9. PL spectr
excitation densitie

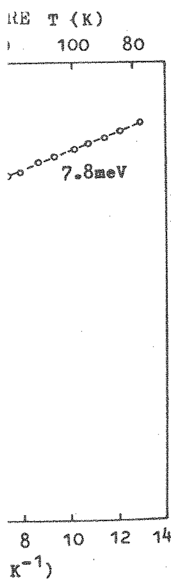
is found that
about 19 me
binding energ
[1]. It means t
the E_s' band.
As seen fro
is increased, f
peak position
the intensity r
increases. It s
of the E_s' banc
E_s band at 77
similarity betv
band at 77 K,
E_s' band at R
recombination
trons in the c
tion energy o
meV, many e
the possibility
relatively smal
the intensity r
decreasing wi
higher levels,
With regard
unlikely to be
the energy ba
tween conduc
would cause t



the near band edge emis-
...e epilayer, excited by the
N_2 laser.

...eless E_s band widens
...avelength and finally
...and E_s' at RT. This
...and is related to free

...ence of the E_s band
...rs on temperature. It



the E_s band intensity from
...layer.

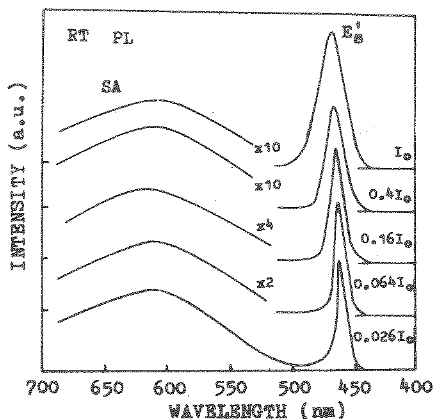


Fig. 9. PL spectra at RT from ZnSe epilayers under various excitation densities, excited by the 337.1 nm line of a N_2 laser with $I_0 = 3$ MW/cm^2.

is found that near RT, the activation energy is about 19 meV, which is in agreement with the binding energy of free excitons in a ZnSe crystal [1]. It means that free excitons are responsible for the E_s' band.

As seen from fig. 9, when the excitation density is increased, first the the E_s' band widens and its peak position shifts towards lower energy; then the intensity ratio of the E_s' band to the SA band increases. It should be noticed that this behavior of the E_s' band at RT is very similar to that of the E_s band at 77 K, as shown in fig. 6. Because of the similarity between the E_s' band at RT and the E_s band at 77 K, it is reasonable to conclude that the E_s' band at RT is attributed to the free exciton recombination following scattering from free electrons in the conduction band. Since the dissociation energy of free excitons in ZnSe is about 20 meV, many excitons are dissociated at RT; thus the possibility of exciton–exciton interactions is relatively small. That is why we could not observe the intensity ratio of the E_s' band to the SA band decreasing with increasing excitation density to higher levels, as mentioned before.

With regard to the origin of the E_s' band, it is unlikely to be due to radiative transitions between the energy band and impurity states [29] or between conduction and valence band [30]. If so, it would cause the E_s' band to shift towards higher energy with increasing excitation density. It is also unlikely to be the I_2 bound exciton due to the small dissociation energy of about 6 meV [31]. In conclusion, the E_s' band at RT in ZnSe epilayers originates from the decay of free excitons after scattering with free electrons in the conduction band.

5. Blue EL

ZnSe MIS diodes using VPE ZnSe epilayers were fabricated. The GaAs face was provided with a Au–Ge alloy ohmic contact. A thick (50–100 nm) insulating layer of ZnSe was evaporated on as-grown or Zn-vapor treated ZnSe epilayers and finally a circular semitransparent Au electrode of 1 mm diameter was evaporated on the top.

Fig. 10 shows the EL spectra of forward biased MIS diodes from Zn-vapor treated (fig. 10a) and as-grown (fig. 10b) ZnSe epilayers. The spectrum at 77 K in fig. 10b has two edge emission bands, E_s at 445.0 nm and FB at 460.0 nm, and two deep center emission bands, D_1 at 540.0 nm and D_2 at 630.0 nm. Let us consider first the two edge emis-

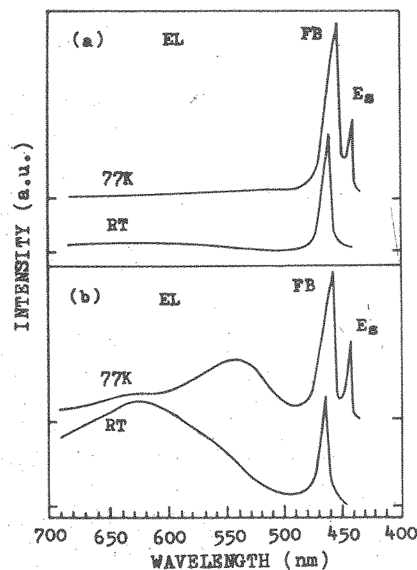


Fig. 10. EL spectra in forward biased ZnSe MIS diodes from Zn-treated (a) and as-grown (b) ZnSe epilayers.

sion bands in fig. 10b. As the temperature is increased, the E_s band broadens and shifts steadily to longer wavelengths, and at RT it lies at 465.0 nm; no self-activated or other deep center emissions are detected. Although the FB band is prominent at 77 K, little trace of the band is detectable at temperatures in excess of 110 K. No trace remained at RT. With increasing temperature, the D_2 band at 630.0 nm increases and its peak position shifts towards shorter wavelength, which indicates that the D_2 band is ascribed to the SA center, but the D_1 band at 540.0 nm becomes weaker and finally disappears at temperatures in excess of 150 K. Comparing fig. 10a with fig. 10b, it is obvious that deep center bands become very weak in fig. 10a. It is worth recording that pure blue EL at RT is observed in forward biased ZnSe MIS diodes from selected Zn treated ZnSe epilayers.

6. Blue stimulated emission

Much work has been done on the stimulated emission from ZnSe under electron beam excitation [32]. There have, however, been several reports on the stimulated emission with photopumping from various ZnSe materials, such as bulk ZnSe single crystal [4], ZnMnSe–ZnSe multiple quantum wells [5,6] and ZnSSe–ZnSe multilayer structures [7]. As far as we are aware, there are no reports of stimulated emission at 77 K excited by the 337.1 nm line of a N_2 laser from VPE ZnSe epilayers.

The ZnSe epilayers on GaAs(100) substrates were cleaved along the natural cleavage plane (110) with widths varying from 100 to 600 μm. These samples were then mounted on holders, such that either front surface emission or emission from a cleaved edge could be observed. According to the method of Shaklee et al. [33], a beam of light of a N_2 laser excites a region of length l on the front surface of the sample. The stimulated emission passes out of the cleaved edge of the sample.

Fig. 11 shows the stimulated emission spectra from the ZnSe epilayers at 77 K at various excitation densities, excited by the 337.1 nm line of a N_2

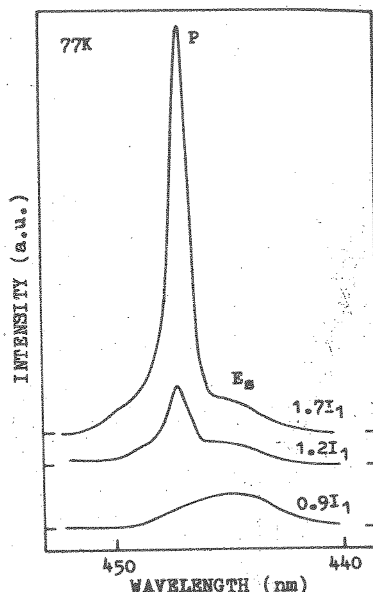
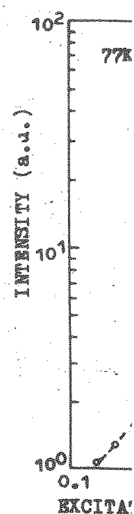


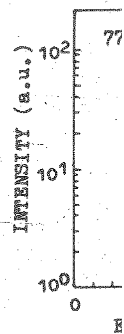
Fig. 11. Stimulated emission spectra at 77 K from VPE ZnSe epilayers, excited by the 337.1 nm line of a N_2 laser (I_1 = 1 MW/cm^2).

laser. At the lowest excitation density of 0.9 MW/cm^2, the spectrum is characterized by only one emission band, E_s, at 445.0 nm [1]. With increasing excitation density, a new band P appears 20 meV below the E_s band, i.e., with an energy separation from the E_s band practically corresponding to the free exciton binding energy of ZnSe (19 meV) [1].

Fig. 12 shows the dependence of PL intensity on pump power for the near band edge emission of a ZnSe epilayer. Three regions can be seen: a region over which the dependence is linear, a region of superlinear dependence and, at the highest excitation densities, a region in which the gain is saturating. As seen in fig. 12, the threshold of 1 MW/cm^2 for the stimulated emission was determined by the clearly observed change from linear behavior to superlinear behavior. In general, the threshold depends on the particular structure investigated as well as on the luminescence energy.

The optical gain was measured according to the method of Shaklee et al. [33]. The method exploits the fact that in the region where stimulated emission predominates, the luminescence intensity I





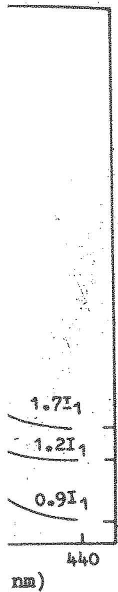


Fig. 12. Dependence of the E_s band intensity on excitation density for a VPE ZnSe epilayer at 77 K.

emitted from the cleaved edge varies exponentially with the excitation lengths l. That is, $I(l)$ is proportional to $\exp(gl)$, where g is the net gain (actual gain minus absorption). Fig. 13 shows the dependence of the PL intensity I on excitation length l in a ZnSe epilayer. The magnitude of the optical gain is found to be about 15 cm^{-1} when the excitation density is 3 MW/cm^2. In our experiment, the 337.1 nm line of a N_2 laser was only used as a pumping source. In this case, the pumping laser photon energy is much larger than the band gap of ZnSe. Moreover, the penetration

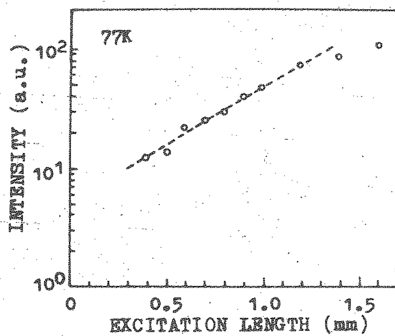


Fig. 13. PL intensity of the P band as a function of excitation length for a VPE ZnSe epilayer at 77 K.

depth is about an order of magnitude shorter than that for resonant pumping. This is why the optical gain obtained here is rather small. Catalano et al. [4] have reported an optical gain of about 300^{-1} cm in ZnSe crystals at 20 K under 2 MW/cm^2 resonant dye laser pumping, but they have not obtained the stimulated emission for ZnSe crystals under the 337.1 line of a N_2 laser. Moreover, they considered that for N_2 laser pumping of ZnSe, a high density of excitons cannot be achieved. We believe that it is possible to obtain suitable gain if the ZnSe epilayers used here are excited by resonant dye laser.

The results mentioned above conclusively demonstrate that stimulated emission occurred from the VPE ZnSe epilayers and originated from the P line, which is associated with exciton–exciton interactions.

7. Conclusions

We have studied the dependence of the optical and electrical properties on growth conditions and obtained high-quality ZnSe epilayers grown on GaAs(100) substrates by the VPE method. A decrease of the intensity ratio of the E_s band to the FB band in VPE ZnSe epilayers has been found under high excitation density. It can be explained by an interaction between excitons. The blue emission at RT from VPE ZnSe epilayers has been ascribed to the decay of free excitons after scattering with free electrons in the conduction band. We have reported for the first time the blue EL at RT in forward biased MIS diodes using VPE ZnSe epilayers and the blue stimulated emission at 77 K from the VPE ZnSe epilayers under the 337.1 nm line of a N_2 laser, which was ascribed to the P band of an exciton–exciton interaction.

Acknowledgements

This work was supported by the National Natural Science Foundation of China. The authors wish to express their thanks to L. Ma, W.Z. Li and J.Y. Zhang for their valuable assistance.

References

[1] X.W. Fan and J. Woods, IEEE Trans. Electron Devices ED-28 (1981) 428.

[2] X.W. Fan and J. Woods, J. Phys. D14 (1981) 1863.

[3] J. Nishizawa, K. Itoh, Y. Okuno and F. Sakurai, J. Appl. Phys. 57 (1985) 2210.

[4] I.M. Catalano, A. Cingolani, M. Ferrara and M. Lugara, Solid State Commun. 43 (1982) 371.

[5] R.B. Bylsma, W.M. Becker, T.C. Bonsett, L.A. Kolodziejski, R.L. Gunshor, M. Yamanishi and S. Datta, Appl. Phys. Letters 47 (1985) 1039.

[6] T.C. Bonsett, M. Yamanishi, R.L. Gunshor, S. Datta and L.A. Kolodziejski, Appl. Phys. Letters 51 (1987) 499.

[7] I. Suemune, K. Yamada, H. Masato, Y. Kan and M. Yamanishi, Appl. Phys. Letters 54 (1989) 981.

[8] T. Yao and S. Maekawa, J. Crystal Growth 53 (1981) 423.

[9] A. Yoshikawa, S. Yamaga, K. Tanaka and H. Kasai, J. Crystal Growth 72 (1985) 13.

[10] R.N. Bhargava, J. Crystal Growth 59 (1982) 15.

[11] P. Lilley, M.R. Czerniak, J.E. Nicholls and J.J. Davies, Phys. Status Solidi (a) 85 (1984) 235.

[12] Sanyo Elec. Co., Ltd., Japan. Kokai Tokkyo Koho Jp. 59-39798.

[13] Sanyo Elec. Co., Ltd., Japan. Kokai Tokkyo Koho Jp. 59-16393.

[14] X.W. Fan, J.Y. Zhang, Z.S. Zhang, S.Y. Wang, A.D. Lu, W.Z. Li, J.X. Jiang and Y.W. Sun, J. Luminescence 31&32 (1984) 957.

[15] J.Y. Zhang and X.W. Fan, Chinese J. Luminescence 8 (1987) 100.

[16] X.W. Fan and J.Y. Zhang, in: Proc. 4th Intern. Workshop on Electroluminescence, Tottori, 1988.

[17] D.Z. Shen and X.W. Fan, Chinese J. Luminescence 9 (1988) 26.

[18] D.Z. Shen and X.W. Fan, in: Proc. 4th Intern. Workshop on Electroluminescence, Tottori, 1988.

[19] Z.K. Tang and X.W. Fan, Chinese J. Luminescence 6 (1985) 314.

[20] X.W. Fan and Z.K. Tang, Chinese J. Luminescence 7 (1986) 336.

[21] Z.K. Tang and X.W. Fan, Chinese J. Luminescence 7 (1986) 348.

[22] G.H. Fan, Q.W. Liao, X.W. Fan, Z.S. Zhang, M. Li and J.Y. Zhang, Chinese J. Rare Metals 8 (1989) 37.

[23] T. Muranoi and M. Furukoshi, Thin Solid Films 86 (1981) 307.

[24] T. Yao, M. Ogura, S. Matsuoka and T. Morishita, Appl. Phys. Letters 43 (1983) 499.

[25] T. Yao, J. Crystal Growth 72 (1985) 31.

[26] S. Fujita, T. Yodo and A. Sasaki, J. Crystal Growth 72 (1985) 27.

[27] K. Yoneda, Y. Hishida, T. Toda, H. Ishii and T. Niina, Appl. Phys. Letters 45 (1984) 1300.

[28] K. Era and D.W. Langer, J. Luminescence 1/2 (1970) 541.

[29] Y. Shirakawa and H. Kukimoto, J. Appl. Phys. 51 (1980) 2014.

[30] S. Fujita, H. Mimoto and T. Noguchi, J. Appl. Phys. 50 (1979) 1079.

[31] M. Yamaguchi, A. Yamamoto and M. Kondo, J. Appl. Phys. 48 (1977) 196.

[32] J.E. Potts, T.L. Smith, H. Cheng, B.J. Yang and B.W. Wessels, J. Crystal Growth 86 (1988) 935.

[33] K.L. Shaklee, R.E. Nahery and R.F. Leheny, J. Luminescence 7 (1973) 384.