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Gain studies and lasing in excitonic waveguides of II–VI submonolayer structures

M. Straßburg[a], N.N. Ledentsov[a,b], A. Hoffmann[a,*], U.W. Pohl[a], D. Bimberg[a], I.L. Krestnikov[b], S.V. Ivanov[b], M.V. Maximov[b], S.V. Sorokin[b], P.S. Kop'ev[b], Zh.I. Alferov[b]

[a] *Institut für Festkörperphysik, Technische Universität Berlin, Hardenbergstr. 36, 10623 Berlin, Germany*
[b] *A.F. Ioffe Physical-Technical Institute, Politechnicheskaya 26, St. Petersburg, 194021, Russian Federation*

Abstract

In this paper we report on gain studies and lasing in excitonic waveguides of CdSe–ZnSe submonolayer structures. In contrast to conventional waveguides and double heterostructure lasers, no significant difference in the *average* refractive index between the cladding and the active layers is necessary, and these regions can be fabricated from the same matrix material. In this approach: (i) the waveguiding effect has a resonant nature and appears on the low-energy side of the strong exciton absorption peak in agreement with the Kramers–Kronig equation (ii) the absorption peak is induced by nanoscale island-like insertions of narrow-gap material in a wide-bandgap matrix. Gain spectra up to 1 kW/cm^2 demonstrate that here the excitonic-waveguide mechanism dominates. At high excitation densities the gain is additionally influenced by the formation of localized biexciton complexes.

Keywords: Excitonic waveguiding; Submonolayer; Gain; II–VI compounds

1. Introduction

Short-wavelength or visible light emitting-(LED) and laser-diodes find applications in many spheres of life. There is a strong competition in this field between devices based on II–VI compounds [1] and on III–V nitrides [2]. Currently, the structure of wide-bandgap lasers is similar to that of conventional infrared III–V lasers, in agreement with the double-heterostructure laser geometry [3]. Thick layers of a wider-bandgap material having a lower refractive index are necessary. However, such a material with a wider bandgap, lattice matched to the active layer, does not always exist, or, if it exists, does not necessarily provide sufficient conductivity. The interest in the shift towards the blue and UV spectral region therefore requires a search of alternative approaches for efficient waveguiding. An attractive idea is to use resonant waveguiding, which originates on the low-energy side of the absorption peak due to a process described by the Kramers–Kronig equation which relates the absorption with the dielectric susceptibility.

The resonant excitonic absorption in II–VI and III–N materials is an attractive candidate for the

* Corresponding author. Fax: +49-30-31422064; e-mail: axel0431@mailszrz.zrz.tu-berlin.de.

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realization of this type of waveguide due to the high exciton binding energies and oscillator strengths as well as the high densities required to screen excitons in these materials (around $10^{19}\,cm^{-3}$) [4]. On the other hand, the exciton-induced lasing, resonant to the range of strongly enhanced refractive index, can hardly be realized in bulk wide-gap II–VI compounds. Free excitons with finite k-values dominating at high excitation densities and observation temperatures cannot recombine radiatively, as was first demonstrated by Gross et al. [5]. More recently, ultrathin submonolayer (SML) [6] or monolayer (ML) [7] insertions were proposed to be used in excitonic waveguides. These insertions proved to form a dense array of nanoscale 2D islands which can efficiently localize excitons. Structures composed of stacked ultrathin insertions result in both lifting of the k-selection rule and a strong increase in the exciton oscillator strength, thus providing a new possibility for lasing and waveguiding in wide-bandgap matrices [8,9].

2. Experimental

Monolayer and submonolayer insertions are most promising for quantum dot (QD) fabrication in II–VI and III–N semiconductors, where the application of excitonic waveguides seems to be most promising. The CdSe submonolayer structures studied here were grown by molecular beam epitaxy (MBE) and consist of a 15 nm thick ZnSe layer grown on a GaAs (1 0 0) substrate, a 1.5 μm thick ZnSSe layer and a 5 nm thick lattice-matched ZnMgSSe lower cladding layer. The active layer is symmetrically confined by ZnSSe layers (50 nm) and represents a 20-period submonolayer superlattice (SML SL) composed of 2/3 ML CdSe insertions separated by 3 nm thick ZnSe layers. The upper ZnMgSSe cladding layer has a thickness of 10 nm and a protective ZnSe layer of 5 nm thickness is grown on top. The investigated II–VI submonolayers were characterized using high-resolution transmission electron microscopy [7]. It was demonstrated that the CdSe deposition results in islands having a lateral size of about 4 nm. The average thickness of the CdSe insertion in the island area is about 1.6 ML and significantly exceeds the *average* thickness of the deposit. Structural characterization of 3D QDs is performed in a number of papers see, e.g., Ref. [10].

To obtain the high excitation density necessary for our investigation we used a dye laser pumped by an excimer laser, providing pulses with a duration of 15 ns at a rate of 60 Hz and maximal pulse energy of up to 1.5 mJ at 440 nm. The SML structures were pumped either by the dye laser at energies below the band edge of the cladding layers or by the excimer laser at 308 nm using similar pulse energies. The samples were mounted in a bath cryostat at 1.8 K or in a helium flow cryostat at temperatures varied between 4 and 300 K. Gain measurements were performed using the stripe length method [11].

3. Results

Photoluminescence (Surface PL), optical reflection (OR) and lasing spectra of the structure are shown in Fig. 1. An intense PL peak is found around 100 meV below the ZnSe bandgap energy. Strong modulation of the optical reflectance is observed in the spectral range resonant with the PL peak. This indicates the high excitonic oscillator strength and, hence, an effective refractive index modulation in the active region. Increase of the exciton oscillator strength in the array of QDs with respect to quantum wells (QWs) has been demonstrated [12]. The resonant enhancement of Δn is 0.15 at the wavelength of the lasing with

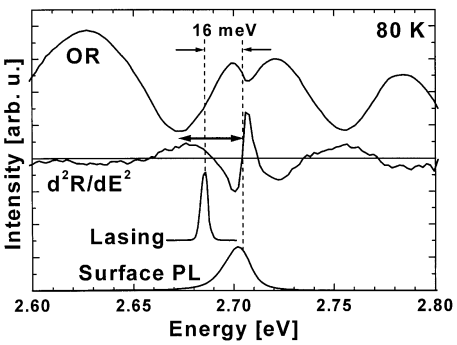

Fig. 1. Photoluminescence (Surface PL), optical reflectance (OR), the second derivative of the OR curve recorded from the top and lasing recorded from the edge of the structure.

respect the refractive index far away from the excitonic absorption energy. It is derived from the fitting of the optical reflectance curve according to the Kramers–Kronig equation. This provides an efficient and complete confinement for photons resonant with the low-energy side of the zero-phonon exciton emission while the contribution to Δn by the average Cd composition (3%) in the SML region is negligible. The antiwaveguiding region is, consequently, placed on the high-energy side of the heavy-hole exciton resonance. To lift the k-selection rule and realize a zero-phonon lasing mechanism [13] in resonance with the exciton waveguiding range [6] we confine the excitons to the CdSe islands formed during the SML growth. The lasing spectrum recorded in the waveguide geometry is 16 meV Stokes shifted with respect to the exciton resonance energy in the OR spectrum. Before we study the gain mechanism using the variable-stripe-length method we investigate the physical processes causing stimulated emissions in CdSe SML.

The dependence of the photoluminescence (edge emission) on excitation densities up to 100 kW/cm^2 is displayed in Fig. 2. The most striking observation is the appearance of a new emission band labeled M around 2.689 eV at 70 kW/cm^2. It exhibits a superlinear growth of emission intensity. Its peak position does not shift with excitation density. At 100 kW/cm^2 the M-band is the dominating feature. The energy difference between the M-band and the free-exciton emission amounts to 11 meV. Comparing the temperature dependence of the M- and free-exciton luminescences at high excitation densities the M-band is the dominating emission at low temperatures while at 30 K the free-exciton luminescence intensity is stronger than that of the M-band. At temperatures higher than 100 K the M- band is quenched. From the Arrhenius plot of the integrated M-band intensity we deduce an activation energy of 9 meV. On the basis of our experimental findings we attribute the M-band to a radiative decay of biexcitons. The determined biexciton binding energy of 9 meV is larger than that of 4 meV found in bulk ZnSe [14]. This is in agreement with observations of Kleinmann [15] who found that localized biexcitons in quantum wells have a binding energy 3–4 times larger than that in the bulk material. In II–VI quantum wells

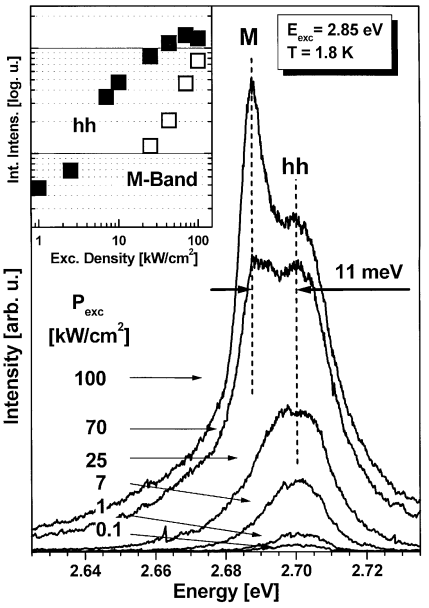

Fig. 2. Edge emission of CdSe SML. With increasing excitation densities the M-band grows at the low-energy side of the free-exciton luminescence. The inset shows the superlinear growth of the biexciton luminescence band.

biexciton binding energies range between 6 meV ZnCdSe [16] and 15 meV [17]. Kreller et al. [17] explained the relatively large biexciton binding energy by the localization of the biexcitons caused by potential fluctuations in a ZnCdSe multi-quantum well.

To identify the gain mechanisms in the CdSe SML we performed gain studies under pulsed excitation using the variable-stripe-length method. Fig. 3 displays a series of gain spectra taken at various excitation densities. Up to densities of 1 kW/cm^2 the gain is determined by the excitonic waveguiding. A fast saturation of the absorption on the low-energy side of the exciton luminescence and the development of a gain peak at an energy 7–10 meV below the exciton resonance in the optical reflectance spectrum of Fig. 1 can be observed. The maximum of the gain here amounts to 105 cm^{-1}. With increasing pump intensity a broadening of the region of optical gain is observed and the gain decreases. Increase in excitation density results in a shift of the gain maximum to the high energy side due to filling of the QD states. At

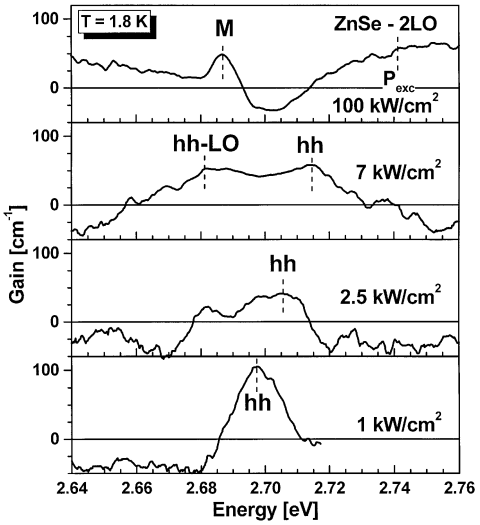

Fig. 3. Gain-spectra of CdSe SML as a function of the excitation density.

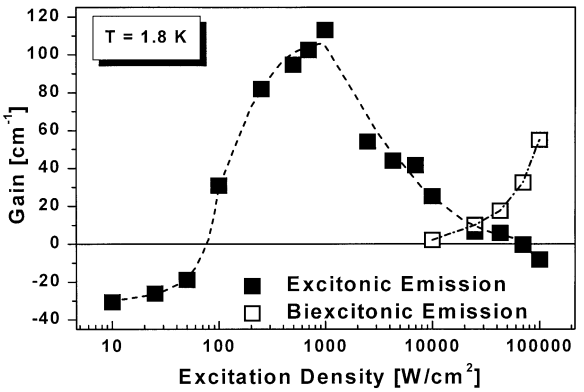

Fig. 4. Gain vs. excitation density in the structure with excitonic waveguide.

excitation densities of 7 kW/cm^2 a gain peak appears at 2.68 eV, which can be explained for example by phonon-assisted processes. This mechanism is in competition with the excitonic recombination whose gain value decreases to 40 cm^{-1}. At excitation densities as high as 100 kW/cm^{-2} the gain curve changes drastically. In the excitonic region a bleaching of the gain is seen and absorption processes dominate. On the high-energy side of the excitonic resonance high density effects in the ZnSSe layer influence the gain behavior. The peak at 2.687 eV denoted M can be attributed to the formation of localized biexcitons as it was shown for ZnCdSe/ZnSe quantum wells in Ref. [18]. The gain of the M-band amounts 55 cm^{-1}.

In Fig. 4 the M-band and heavy-hole exciton gain are plotted as functions of the excitation density. The onset of the gain due to the M-band emission is at a P_{exc} of 10 kW/cm^2. With increasing excitation density P_{exc} its gain value rises up to 55 cm^{-1} and dominates. At low P_{exc} a fast saturation of the absorption on the low-energy side of exciton luminescence with P_{exc} is demonstrated. The monotonic increase in gain with P_{exc} up to values of 100 cm^{-1} and the gain saturation (see Fig. 4) as well as the appearance of a dip in the gain curve upon a further increase of P_{exc} are clearly visible. For even higher P_{exc} this dip develops to an absorption peak since, after the excitonic absorption is completely saturated, no waveguiding is possible anymore, and the emitted light is absorbed by the GaAs substrate. This intriguing gain suppression effect provides unique possibilities for a new generation of devices based on optical bistability and for the generation of ultrashort pulses.

4. Conclusion

Excitonic waveguides represent a unique system with a high device potential, particularly in the ultraviolet spectral region. The recent progress became possible due to the application of self-organized growth of quantum islands by using submonolayer and monolayer deposition in the CdSe-(Zn,Mg)(S,Se) materials system.

Acknowledgements

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References

[1] M.A. Haase, J. Qui, J.M. de Puydt, H. Cheng, Appl. Phys. Lett. 59 (1991) 1272.

[2] S. Nakamura, MRS Internet J. Nitride Semiconductor Res. 2 (1997) 5.
[3] Zh.I. Alferov, Fizika i Tekn. Poluprovodn. 1 (1967) 436.
[4] Zh.I. Alferov, S.V. Ivanov, P.S. Kop'ev, A.V. Lebedev, N.N. Ledentsov, M.V. Maximov, I.V. Sedova, T.V. Shubina, A.A. Toropov, Superlattices and Microstructures 15 (1994) 65.
[5] E. Gross, S. Permogorov, A. Razbirin, J. Phys. Chem. Solids 27 (1966) 1647.
[6] N.N. Ledentsov, I.L. Krestnikov, M.V. Maximov, S.V. Ivanov, S.L. Sorokin, P.S. Kop'ev, Zh.I. Alferov, D. Bimberg, C.M. Sotomayor Torres, Appl. Phys. Lett. 69 (1996) 1343.
[7] N.N. Ledentsov, I.L. Krestnikov, M.V. Maximov, S.V. Ivanov, S.L. Sorokin, P.S. Kop'ev, Zh.I. Alferov, D. Bimberg, C.M. Sotomayor Torres, Appl. Phys. Lett. 70 (1996) 2766.
[8] I.L. Krestnikov, M.V. Maximov, S.V. Ivanov, N.N. Ledentsov, S.V. Sorokin, A.F. Tsatsul'nikov, O.G. Lyublinskaya, B.V. Volovik, P.S. Kop'ev, C.M. Sotomayor Torres, Semiconductors 31 (1997) 127; Fiz. Tekh. Poluprovodn. 31 (1997) 230.
[9] I.L. Krestnikov, M.V. Maximov, S.V. Ivanov, S.L. Sorokin, S.A. Permogorov, A.N. Reznitsky, A.V. Kornievski, N.N. Ledentsov, D. Bimberg, C.M. Sotomayor Torres, in: M. Scheffler, R. Zimmermann (Eds.), Proc. 23rd Int. Conf. on the Physics of Semiconductors, vol. 1, Berlin, Germany, July 21–26, 1996, World Scientific, Singapore, 1996, p. 3187.
[10] For a review see e.g. N.N. Ledentsov, M. Grundmann, N. Kirstaedter, O. Schmidt, R. Heitz, J. Böhrer, D. Bimberg, V.M. Ustinov, V.A. Shchukin, P.S. Kop'ev, Zh.I. Alferov, S.S. Ruvimov, A.O. Kosogov, P. Werner, U. Richter, U. Gösele, J. Heydenreich, in Proc. MSS7, Madrid, 1995, Solid State Electron. 40 (1996) 785.
[11] C. Benoit a la Guillaume, J.M. Denber, F. Salvan, Phys. Rev. 177 (1969) 567.
[12] M.V. Belousov, N.N. Ledentsov, M.V. Maximov, P.D. Wang, I.N. Yassievich, N.N. Faleev, I.A. Kozin, V.M. Ustinov, P.S. Kop'ev, C.M. Sotomayor Torres, Phys. Rev. B 51 (1995) 14346.
[13] J. Ding, H. Jeon, T. Ishihara, M. Hagerott, A.V. Nurmikko, H. Luo, N. Samarth, J. Furdyna, Phys. Rev. Lett. 69 (1992) 1707.
[14] V. Kutzer, B. Lummer, R. Heitz, A. Hoffmann, I. Broser, E. Kurtz, D. Hommel, J. Crystal Growth 159 (1996) 776.
[15] D.A. Kleinmann, Phys. Rev. B 28 (1983) 871.
[16] H. Grempel, A. Diessel, W. Ebeling, J. Gutowski, K. Schüll, B. Jobst, D. Hommel, M.F. Pereira Jr., K. Henneberger, Phys. Stat. Sol. (b) 194 (1996) 199.
[17] F. Kreller, J. Puls, F. Henneberger, Appl. Phys. Lett. 69 (1996) 3406.
[18] F. Kreller, M. Lowisch, J. Puls, F. Henneberger, Phys. Rev. Lett. 75 (1995) 2420.