

## Gain studies of (Cd, Zn)Se quantum islands in a ZnSe matrix

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By inserting stacked sheets of nominally 0.7 monolayer CdSe into a ZnSe matrix we create a region with strong resonant excitonic absorption. This leads to an enhancement of the refractive index on the low-energy side of the absorption peak. Efficient waveguiding can thus be achieved without increasing the average refractive index of the active layer with respect to the cladding. Processed high-resolution transmission electron microscopy images show that the CdSe insertions form Cd-rich two-dimensional (Cd, Zn)Se islands with lateral sizes of about 5 nm. The islands act as quantum dots with a three-dimensional confinement for excitons. Zero-phonon gain is observed in the spectral range of excitonic and biexcitonic waveguiding. At high excitation densities excitonic gain is suppressed due to the population of the quantum dots with biexcitons. © 1998 American Institute of Physics. [S0003-6951(98)03008-3]

II–VI semiconductors are promising materials for short-wavelength laser diodes. For efficient waveguiding, the common double heterostructure geometry<sup>1</sup> of such wide-bandgap lasers requires thick layers of a wider-bandgap material which has a lower refractive index, a sufficient conductivity and is lattice-matched to the active region. Such cladding materials do, however, not exist for all spectral ranges and all possible active layer materials. Furthermore, since resonant emission from excitons is only possible at  $k=0$ ,<sup>2</sup> phonons or other inelastic scattering processes are required to initiate lasing at high excitation densities and high temperatures. This leads to a reduction of the gain. Recently, the alternative concept of vertically stacked quantum dots inserted into the active layer has been introduced. Here, zero-phonon lasing in the spectral region of excitonic waveguiding can be realized without thick cladding layers of lower refractive index.<sup>3,4</sup> In such structures the material gain is increased by a giant oscillator strength in the excitonic region.<sup>5</sup> The waveguiding is based on a large refractive index enhancement ( $\Delta n$ ) on the low energy side of excitonic absorption.<sup>6</sup> In our work the investigation of the structural properties provides a clear proof of island formation due to submonolayer CdSe insertions. In a structure with stacked insertions, efficient excitonic gain is found resulting from localization at the islands which act as electronic quantum dots. At high excitation densities an appearance of biexcitonic gain at the expense of excitonic gain is observed.

The structures were grown by molecular beam epitaxy<sup>7</sup> on semi-insulating GaAs substrate after deposition of 20 nm ZnSe and 1.35  $\mu\text{m}$  ZnS<sub>0.06</sub>Se<sub>0.94</sub> buffer layers. The active regions consist of a single sheet (structure A) or a twenty-

period superlattice (structure B) composed of nominally 0.7 monolayer (ML) CdSe insertions separated by 30 Å ZnSe. The active regions were symmetrically confined by 50 nm ZnSSe layers and 10 nm lattice-matched (Zn,Mg)(S,Se) barriers. For the high-resolution transmission electron microscopy (HRTEM) investigations we also fabricated an eight-period superlattice structure with nominal CdSe thickness of 1.1 ML (structure C).

Cross-section HRTEM was performed along the  $\langle 110 \rangle$ -direction using a PHILIPS CM 200 FEG/ST electron microscope with a Scherzer resolution of 0.24 nm. Gain spectra were recorded at 1.8 K by the variable-stripe length method<sup>8</sup> using a dye laser. The pulse duration was 15 ns providing a maximum pulse power of 100  $\text{kW}/\text{cm}^2$  at 2.8 eV.

Figure 1(a) shows an  $\langle 110 \rangle$ -HRTEM image of the bottom part structure C containing three pairs of CdSe–ZnSe layers. To reveal the distribution of the cadmium, digitized HRTEM images were processed with the evaluation program DALI,<sup>9</sup> which allows the determination of local lattice parameters (LLP). The chemical composition was deduced on the basis of Vegard's law assuming a linear dependence of the Cd content on the CdZnSe lattice parameter and a coherently strained structure. A two-dimensional lattice model was generated with a reference region in the ZnSe layer at the bottom of the image. In Fig. 1(b) we show a color-coded map of the LLP in growth direction,  $a_{\perp}$ , measured relative to  $a_{\perp}$  of the ZnSe reference lattice. The green, yellow, and red areas indicate layers with a larger local  $a_{\perp}$ . Since CdSe has a larger bulk lattice parameter (6.081 Å) than ZnSe (5.6697 Å) these layers are identified as (Cd,Zn)Se. Figure 1(b) shows that the deposition of about 1ML CdSe leads to the formation of (Cd,Zn)Se layers which are not homogeneous. Regions of increased Cd content with a lateral size of 40–50 Å can be observed. The contrast modulation in the

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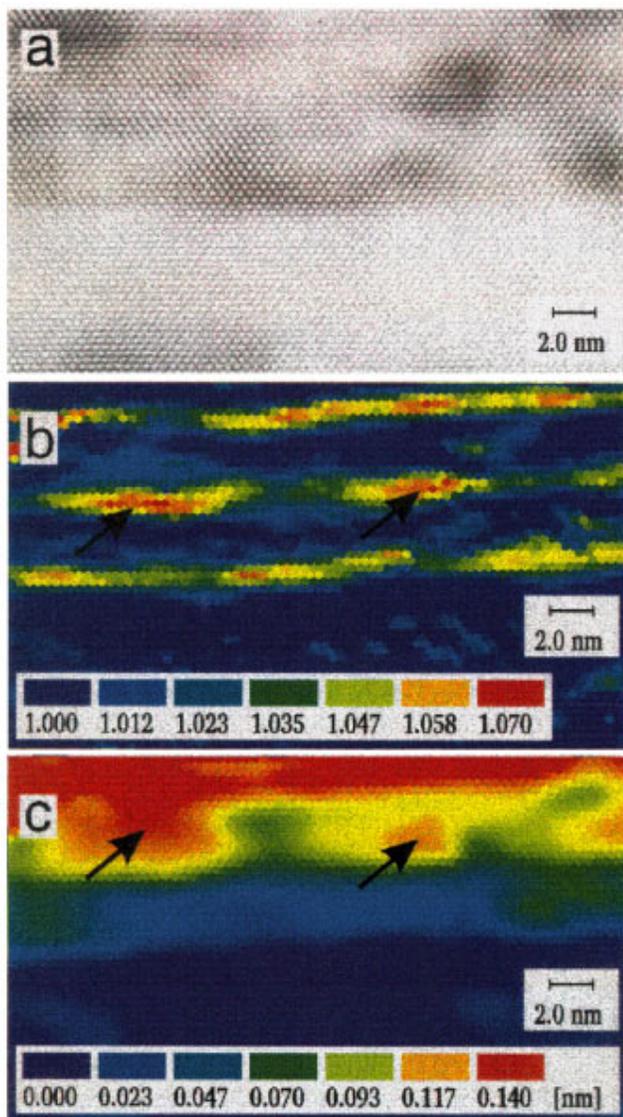


FIG. 1. HRTEM image of structure C showing 3 cycles of the CdSe–ZnSe superlattice (a). Color-coded maps of the local lattice parameter  $a_{\perp}$  along the vertical direction (b) and the total atom displacements with respect to the underlying ZnSe lattice (c) for the same area.

vertical direction due to the (Cd, Zn)Se insertion corresponds to 4 MLs. This is an upper limit because steps in the imaging direction may also lead to a contrast broadening. The average CdSe composition in the Cd-rich islands amounts to 40% if 4 ML average thickness is assumed. The Cd content in the (Cd,Zn)Se layer between the islands is at least twice lower.

Figure 1(c) shows a color-coded map of the total atomic displacements in the vertical [001]-direction for the same region. The regions with increased displacements are located directly above the regions with larger Cd content (see arrows). The effect originates from a bowing of the (001)-lattice planes in the [001] direction which is caused by the underlying regions of increased tetragonal distortion. This is a further indication of the significant local Cd enhancement. The HRTEM data of structure B are similar to those described above.

The formation of CdSe-rich islands observed in HRTEM leads to a strong photoluminescence (PL). Figure 2 shows a comparison between the stimulated edge emission and the

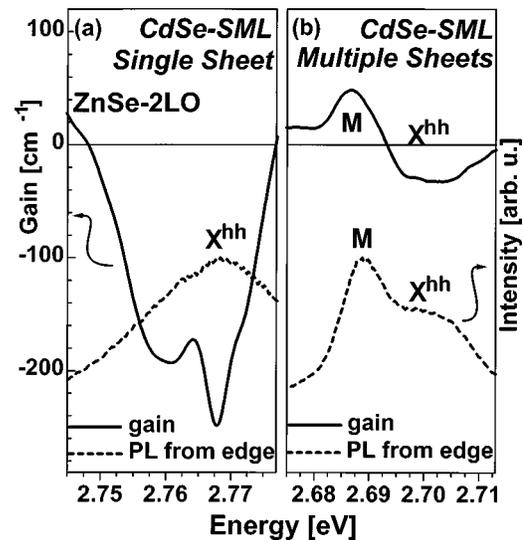


FIG. 2. Gain and stimulated edge emission spectra of a CdSe/ZnSe submonolayer sheet (a) and a submonolayer superlattice (b) recorded at 1.8 K with an excitation density of  $100 \text{ kW/cm}^2$ . Strong absorption appears in the excitonic range of the single sheet (a) whereas the superlattice (b) exhibits strong zero-phonon gain at the excitonic M band.

gain of sample A [Fig. 2(a)] and the superlattice sample B [Fig. 2(b)]. The luminescence of the heavy hole exciton ( $X^{\text{hh}}$ ) localized at the CdSe islands appears at 2.768 eV and at 2.700 eV for samples A and B, respectively. Due to the strong localization the islands act as electronic quantum dots. Sample B shows at high excitation densities a new, dominating luminescence line ( $M$  line) at 2.689 eV on the lower-energy side of the excitonic emission. No intensity-dependent redshift of this line occurs, like for electron-hole plasma recombination. We assign this line to the recombination of biexcitons localized in the quantum dots having a binding energy of about 11 meV. This is consistent with the values published for biexcitons in ZnCdSe-based quantum well systems, ranging from 6 meV<sup>10</sup> to 15 meV.<sup>11</sup> Large biexciton binding energies are attributed to localization effects on potential fluctuations in ZnCdSe multiquantum well systems.<sup>11</sup>

The gain spectra of samples A and B differ strongly. The large absorption ( $250 \text{ cm}^{-1}$ ) in the energy range of the  $X^{\text{hh}}$  emission observed for the single sheet sample A is due to the insufficient thickness of the region with a refractive index modulation providing no waveguiding. Thus, the emitted light penetrates to the GaAs substrate and is absorbed there. Consequently no excitonic or biexcitonic gain is found in this sample. For a superlattice we have demonstrated in previous work<sup>12</sup> that a strong modulation of the refractive index due to excitonic absorption leads to efficient waveguiding.

While a weak absorption occurs at the transition energy of the  $X^{\text{hh}}$  emission, an efficient gain of  $55 \text{ cm}^{-1}$  is observed at the spectral position of the  $M$  line. Consequently, a zero-phonon gain induced by biexcitons must be concluded. The high efficiency results from breaking the  $k$ -selection rule at the quantum dots due to electron-hole pair localization. In this case no LO phonons are necessary to accommodate a high in-plane  $k$  vector of excitons.

The effect of excitonic waveguiding can be demonstrated by the excitation density dependent evolution of the

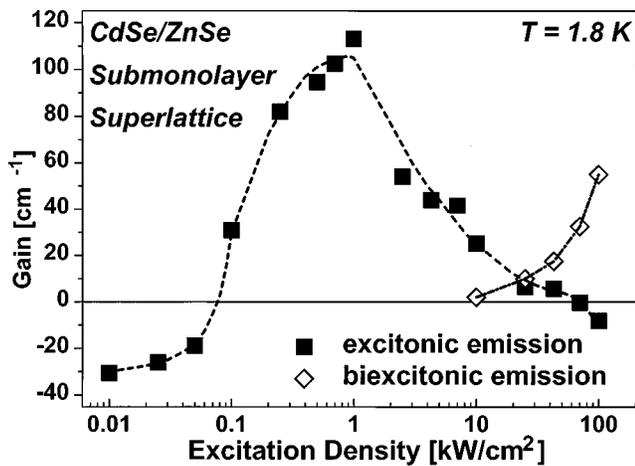


FIG. 3. Excitation density-dependent gain of the submonolayer superlattice recorded at the excitonic and biexcitonic emission  $X^{\text{hh}}$  and  $M$ , respectively. Gain saturation depressing excitonic waveguiding occurs at medium excitation densities. A superlinear increase in biexcitonic gain is observed at high excitation densities at the expense of excitonic gain.

gain for the  $X^{\text{hh}}$  exciton and the biexciton  $M$  line (Fig. 3). At lowest excitation densities absorption occurs due to a low exciton density and a resulting minor modulation of the refractive index. The lack of waveguiding leads to a loss of the generated light from the active region into the substrate. At higher excitation densities, the gain increases gradually up to  $100 \text{ cm}^{-1}$  as a consequence of sufficient refractive index modulation, followed by a saturation at excitation densities up to  $70 \text{ kW/cm}^{-2}$ . At high excitation densities all quantum dots become populated with excitons and a further increase of the excitation density results in their conversion to biexcitons emitting at lower photon energies. The excitonic gain, thus, decreases due to a lack of quantum dots populated with excitons.<sup>13</sup> Consequently the modulation of the refractive index is reduced, waveguiding starts to disappear and absorption to the GaAs substrate is increased. At excitation densities of  $100 \text{ kW/cm}^{-2}$  optical losses exceed the material gain and only absorption is detected.

In conclusion, we have demonstrated that sheets of nominally 0.7 ML CdSe inserted into a ZnSe matrix form two-dimensional Cd-rich (Zn, Cd)Se islands. The islands have 4 ML average thickness, about 5 nm lateral size and can be regarded as electronic quantum dots. In a sample with a

sufficient number of stacked sheets, excitons localized at the quantum dots induce an enhancement of the refractive index leading to efficient waveguiding and zero-phonon excitonic gain. Biexcitonic gain appears at high excitation density at the expense of the excitonic gain.

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- <sup>1</sup>Zh. I. Alferov and R. F. Kazarinov, Double Heterostructure Laser, Authors Certificate No. 27448, No. 950840 with a priority from 30 March, 1963.
- <sup>2</sup>E. Gross, S. Permogorov, and A. Razbirin, *J. Phys. Chem. Solids* **27**, 1647 (1966).
- <sup>3</sup>N. N. Ledentsov, I. L. Krestnikov, M. V. Maximov, S. V. Ivanov, S. L. Sorokin, P. S. Kop'ev, Zh. I. Alferov, D. Bimberg, and C. M. Sotomayor Torres, *Appl. Phys. Lett.* **69**, 1343 (1996).
- <sup>4</sup>I. L. Krestnikov, M. V. Maximov, S. V. Ivanov, S. L. Sorokin, S. Permogorov, L. N. Reznitsky, A. V. Kornievski, N. N. Ledentsov, D. Bimberg, and C. M. Sotomayor Torres, in *Proceedings of the 23th International Conference on the Physics of Semiconductors*, edited by M. Scheffler and R. Zimmermann (World Scientific, Singapore, 1996), Vol. 4, p. 3187.
- <sup>5</sup>M. V. Belousov, N. N. Ledentsov, M. V. Maximov, P. D. Wang, I. N. Yasievich, N. N. Faleev, I. A. Kozin, V. M. Ustinov, P. S. Kop'ev, and C. M. Sotomayor Torres, *Phys. Rev. B* **51**, 14346 (1995).
- <sup>6</sup>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, and A. A. Toropov, *Superlattices Microstruct.* **15**, 65 (1994).
- <sup>7</sup>S. V. Ivanov, S. V. Sorokin, P. S. Kop'ev, D. R. Kim, H. D. Jung, and H. S. Park, *J. Cryst. Growth* **159**, 16 (1996).
- <sup>8</sup>Benoit a la Guillaume, C. Debever, and J. Salvan, *Phys. Rev.* **177**, 567 (1969).
- <sup>9</sup>A. Rosenauer, S. Kaiser, T. Reisinger, J. Zweck, W. Gebhardt, and D. Gerthsen, *Optik* **102**, 63 (1996).
- <sup>10</sup>H. Gempel, A. Diessel, W. Ebeling, J. Gutowski, K. Schüll, B. Jobst, D. Hommel, M. F. Pereira, Jr., and K. Henneberger, *Phys. Status Solidi B* **194**, 199 (1996).
- <sup>11</sup>F. Kreller, J. Puls, H.-J. Wünsche, and F. Henneberger Torres, in *Proceedings of the 23th International Conference on the Physics of Semiconductors*, edited by M. Scheffler and R. Zimmermann (World Scientific, Singapore, 1996), Vol. 3, p. 2111.
- <sup>12</sup>I. L. Krestnikov, S. V. Ivanov, P. S. Kop'ev, N. N. Ledentsov, S. V. Sorokin, and D. Bimberg, 2nd International Conference on Low Dimensional Structures and Devices, Lisbon, Portugal, 1997, accepted for publication in *Mater. Sci. Eng. B: Solid State Materials for Advanced Technology*.
- <sup>13</sup>M. Grundmann and D. Bimberg, *Jpn. J. Appl. Phys., Part 1* **36**, 4181 (1997).