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Gain to absorption conversion by increasing excitation density in excitonic waveguides

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Abstract

Exciton waveguiding was proposed to improve optical confinement of II–VI lasers. Strong resonant exciton absorption induces a refractive index enhancement (Δn) on the low-energy side of the absorption peak. By stacking sheets of submonolayer (SML) CdSe insertions into a ZnSe matrix, we fabricated a region in ZnSe with strong resonant exciton absorption. In this region, the k-selection rule is lifted due to exciton localization by the SML islands. Therefore, a zero-phonon lasing mechanism resonant to the exciton waveguiding region could be realized. Using the variable-stripe-length method, we study the intensity-dependent optical gain of the ZnSe/CdSe submonolayer superlattices in a $ZnS_{0.06}Se_{0.94}$ matrix. © 1998 Elsevier Science B.V. All rights reserved.

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1. Introduction

II–VI compounds are promising materials for growing short-wavelength LED's and laser diodes. The common structure of such wide-band gap lasers is based on the double heterostructure geometry as well known from conventional III–V lasers [1]. Therefore, thick layers lattice-matched to the active region with a lower refractive index are required for efficient waveguiding. However, the production of this material with a wider band gap providing sufficient conductivity and suitable lattice parameters has not been achieved in sufficient quality. Additionally, it was shown by Gross et al. [2] that resonant emission from excitons is still possible at $k = 0$, so phonons or other inelastic

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scattering processes are required to initiate lasing at high excitation densities and observation temperatures. To avoid these problems, recently, the concept of vertically ordered quantum dots was introduced. Here, zero-phonon lasing resonant to the exciton waveguiding region was realized [3]. As a result of the giant oscillator strength in the excitonic region, the material gain is increased and the threshold current density is reduced significantly [4]. In our work, we investigated the intensity and temperature-dependent gain behavior of ZnSe/CdSe submonolayer superlattices in a $ZnS_{0.06}Se_{0.94}$ matrix.

2. Experimental

The samples are grown with the Stranski–Krastanov method as described in detail in Ref [5]. The investigated structures consist of a 20 nm thick ZnSe buffer grown on semi-insulating GaAs substrate using MBE, and a 350 nm $ZnS_{0.06}Se_{0.94}$ layer. The lower cladding layer was grown as lattice-matched ZnMgSSe, the active region consists of a 20-period submonolayer superlattice composed of 1/3 monolayer CdSe insertions separated by 30 Å ZnSe layers and symmetrically confined by ZnSSe layers, as shown in Fig. 1.

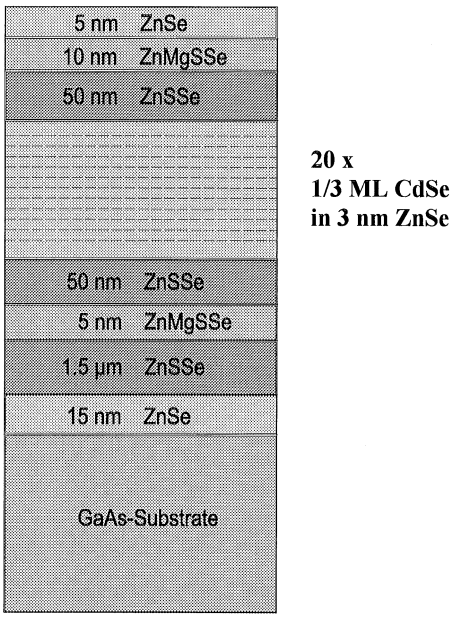

Fig. 1. Scheme of the investigated sample structure. The active region consist of 20-period superlattice of 1/3 monolayer (ML) CdSe inserted 3 nm ZnSe matrix.

The gain spectra are recorded using the variable-stripe-length method. In order to get sufficient excitation densities, we use a pulse laser system, consisting of a dye laser pumped by an excimer laser. The pulse duration is 15 ns at a rate of 30 Hz, providing maximum pulse energies of 1.5 mJ at 440 nm. The spectra are recorded at 1.8 K.

3. Results

For a detailed elucidation of the observed gain mechanisms the features in the stimulated emission characteristics have to be discussed first. Fig. 2 shows a comparison between the spontaneous emission from the surface (Fig. 2a) and the stimulated emission from the edge (Fig. 2b). The luminescence at 2.703 eV originates from the recombination of the X^{hh}- exciton localized at CdSe islands, as was clearly proven in Ref. [6]. The stimulated emission of the X^{hh}-exciton is red-shifted to about 3 meV induced by strong reabsorption processes on the high-energy tail of the excitonic emission [7].

Additionally, the observed luminescence band labeled as M on the lower energy side of the exciton at 2.689 eV becomes dominant in the stimulated emission. From the nonlinear increase of this luminescence band in intensity-dependent measurements (Fig. 3), we assign this band to the recombination of biexcitons. Since no intensity-dependent red shift of the energetic position occurs, we can exclude plasma recombination as the underlying physical process.

From the temperature-dependent development of the integrated M-band intensity, we can deduce a biexciton binding energy of about 9 meV. This is consistent with values published for biexcitons in ZnCdSe-based quantum well systems, ranging between 6 meV [8] and 15 meV [9]. Kreller et al. [10] explained the relatively large biexciton binding energy in terms of localization effects on potential

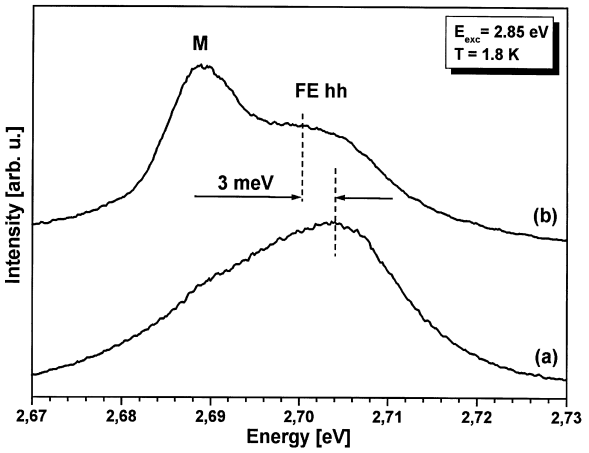

Fig. 2. Low-temperature photoluminescence spectra of a ZnSe–CdSe-SML-superlattice structure recorded at high excitation densities using an excimer-laser pumped dye-laser. (a) shows the spontaneous emission from the surface attributed to free excitons (FE hh) localized at CdSe islands. The stimulated emission is labeled with (b). Additional to the excitonic recombination, we observe luminescence attributed to biexcitons (M band).

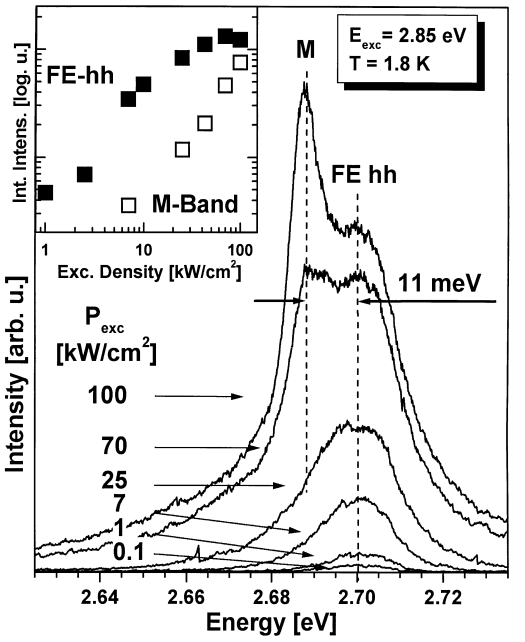

Fig. 3. Intensity-dependent spectra of the stimulated emission at 1.8 K. The inset shows the superlinear increase of the M-band and the saturation of the excitonic emission.

fluctuations in a ZnCdSe multi-quantum well system.

Fig. 4 displays a series of gain spectra taken at various excitation densities. Up to 1 kW/cm^2 the observed gain is resonant to the X^{hh} exciton, demonstrating the effect of excitonic waveguiding. The gain maximum amounts to 105 cm^{-1}. With increasing excitation densities, phonon-assisted gain is observed and an additional gain peak labeled with B develops at 2.71 eV. From luminescence experiments [11] it is known that this effect is correlated to an increased number of dots with reduced size contributing to the luminescence. These mechanisms are in competition with the X^{hh} recombination, whose gain value decreases to 40 cm^{-1}. At highest excitation densities the spectral shape of the gain is drastically changed. In the excitonic region absorption is observed, whereas the gain is dominated by biexcitonic recombination and contributions of the ZnSSe layers. The maximum value for the M band is 55 cm^{-1}.

Fig. 5 shows the intensity-dependent development of the gain for the X_{hh} exciton and the M band, respectively. The onset of the gain induced by biexcitons is at excitation densities P_{exc} of 10 kW/cm^{-1}. At lowest excitation densities, a fast saturation of the absorption on the low-energy side of the excitonic luminescence is observed. Subsequently, the gain increases steadily up to 100 cm^{-1}, followed by a saturation up to 70 kW/cm^{-1}. This saturation can be seen as a dip in the gain curve on the excitonic resonance (see Fig. 3). For even higher excitation densities this dip develops into an absorption peak since, after the excitonic absorption is completely saturated, the free carriers induce a significant change of the refractive index and therefore no excitonic waveguiding is possible. In this case the emitted light is absorbed by the GaAs substrate.

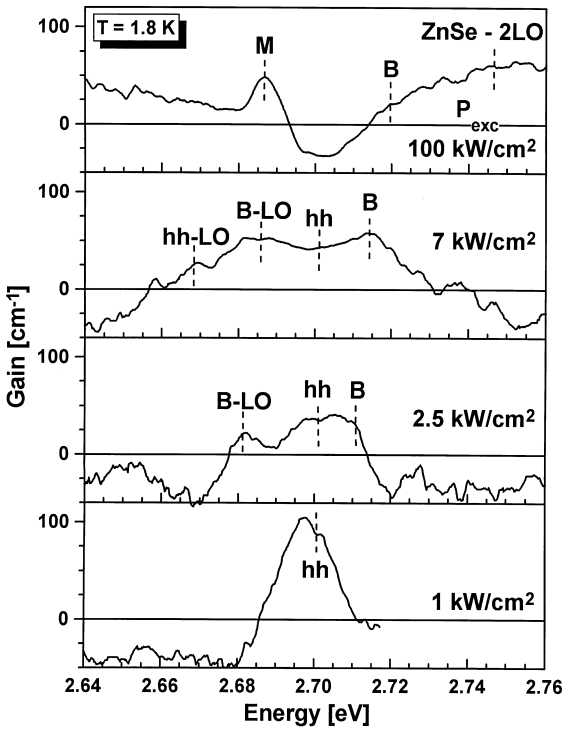

Fig. 4. Intensity-dependent gain spectra of the excitonic region in CdSe/ZnSe SML superlattice. At low excitation densities, the gain is slightly red-shifted to exciton absorption induced by self-absorption effects.

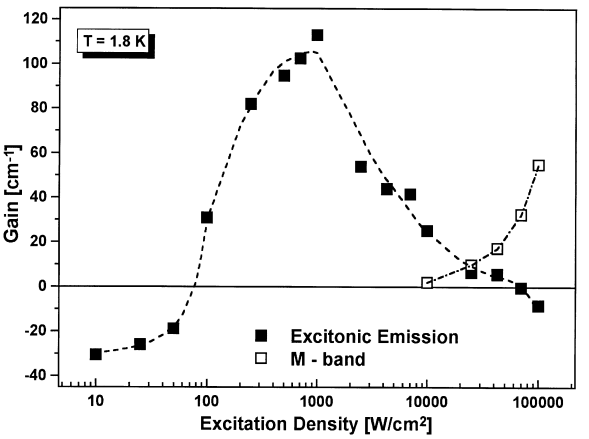

Fig. 5. At low excitation densities excitonic absorption corresponding to the low-energy side of the PL line saturates and gain peak develops at this energy. With increasing P_{Exc}, excitonic absorption is completely saturated and the free excess carriers cannot relax in the excitonic states further. As shown by Fig. 4, at highest excitation densities the free carriers depressing the excitonic waveguiding recombine in the barriers leading to the observed broad gain emission outside the excitonic range of the CdSe islands. Additionally, a superlinear increase in biexcitonic gain is observed at excitation densities larger than 25 kW/cm^2.

4. Conclusion

In this paper we reported on gain mechanisms of ZnSe/CdSe submonolayer superlattices in a $ZnS_{0.06}Se_{0.94}$ matrix. At low excitation densities the maximum gain is observed 2.2 meV below the excitonic absorption, originating from localized excitons on CdSe islands. From intensity-dependent measurements, we found gain to absorption conversion induced by high carrier densities in the barrier materials, leading to an index bleaching and reduced excitonic waveguiding. Additionally, at higher excitation densities biexcitonic recombination dominates the gain spectra with a maximum value of 55 cm^{-1}.

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