MBE grown high-quality CdSe-based islands and quantum wells using CdS compound and Se

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Abstract

Using CdS compound and elemental Se, we have grown CdSe/ZnSe quantum structures with improved optical and structural properties exploiting an exchange reaction which leads to the substitution of sulfur by selenium. Typical S contamination is below 2%. A possibly enhanced surface diffusion of adatoms caused by the high CdS oven temperature and a surfactant-like effect of the S–Se exchange lead to a suppression of Cd segregation in the case of migration enhanced epitaxy with long Se exposure times. The new growth method leads to CdSe quantum wells with outstanding optical quality. Their properties are compared to CdSe island structures obtained in a non-migration-enhanced growth mode. © 2001 Elsevier Science B.V. All rights reserved.

1. Introduction

Self-organized quantum island structures are widely investigated because of their potential for opto-electronic applications. In particular, the enhancement of the oscillator strength within a small spectral range observed in quasi-zero-dimensional systems is desired to improve the lasing threshold. This demands an improvement in the controllability and overall quality of such structures. While in the III–V system island formation is already established [1], II–VI semiconductor-based structures suffer from a strong tendency of Cd segregation and CdSe/ZnSe intermixing, which hinders a classical Stranski–Krastanow type islanding [2,3]. However, CdSe/ZnSe-based systems would be the most suitable candidates for opto-electronic devices in the green spectral range. In this report, we will show that the introduction of sulfur during molecular beam epitaxy (MBE) of CdSe by using a CdS compound source as Cd supply instead of the conventional Cd elemental

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source strongly enhances the overall quality of such structures.

2. Experimental procedure

The samples were grown by MBE on (0 0 1) GaAs wafers using elemental Zn and Se for ZnSe buffer growth at 280°C. All growth processes were monitored by in situ reflection high-energy electron diffraction (RHEED). CdSe-based quantum wells and islands were obtained by inserting thin CdSe layers obtained by a substitutional reaction of CdS compound with elemental Se which leads to an almost complete replacement of sulfur by selenium. Optical measurements indicate a sulfur contamination below 2% which is corroborated by the lattice constant obtained for thick CdSe epilayers [4].

Fig. 1(a) shows the growth sequence for migration-enhanced growth with periodic shutter cycles which was used to obtain quantum wells (QW) of outstanding quality, represented here by samples 1 and 3. In order to reduce Cd segregation, lengthy Se exposure times of 30 s were chosen after a deposition of nominally $\frac{1}{2}$ monolayer (ML) CdSe, gauged according to the RHEED oscillations observed during continuous exposure as shown in Fig. 1(b). There are several advantages to this method. The rather high CdS oven temperature of 650°C improves the surface diffusion of adatoms.

Furthermore, an effect similar to a self-surfactant effect which seems to be caused by the exchange reaction between sulfur and Se plays an important role. Further results can be found elsewhere [4,5].

3. Results and discussion

Fig. 2. shows the photoluminescence (PL) spectra of two representative samples. Sample 1, Fig. 2(a) is a QW with a nominal deposition of $\sim$2.5–3 monolayers (ML) based on the observation of half a ML oscillation per deposition cycle by RHEED, see Fig. 1.

Sample 2, Fig. 2(b), contains a layer of nominally $\sim$2 ML CdSe which have been reformed into Cd-enriched islands inserted in a broadened, intermixed wetting layer with a width of 10 ML and an average Cd concentration of $\sim$15%. Two types of islands can be distinguished by transmission electron microscopy (TEM) [5]. Firstly, natural type A islands (cf. Ref. [6]), constitute more random Cd concentration fluctuations (up to 30%) with a typical extension of 3 nm and rather high densities of more than 10^{11} cm^{-2}. Secondly, self-organized islands of type B have a higher Cd concentration of up to 60%, diameters of typically 5 nm and a density of $\sim$5 × 10^{10} cm^{-2}. The presence of these islands mirrors directly in the high number of sharp discrete lines observed in the micro-PL. The shape and the distribution of lines
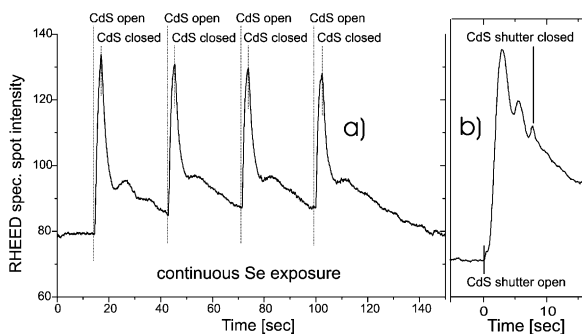

Fig. 1. RHEED oscillations observed during CdSe deposition with CdS and Se exposures. (a) Sequence for migration enhanced epitaxial (MEE) growth of a quantum well. (Here samples 3 and 1 were grown with 6 cycles.) (b) RHEED oscillations for about 2.3 ML CdSe deposited for sample 2.
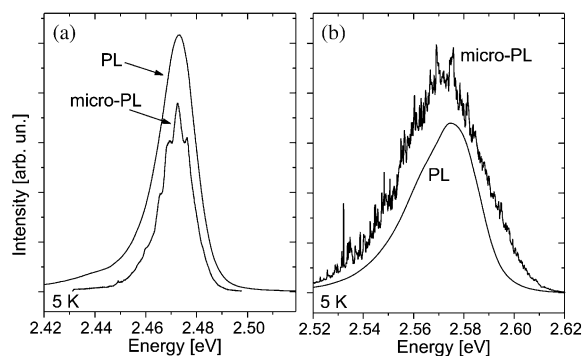

Fig. 2. PL and micro-PL spectra for two representative samples discussed in the text. (a) Six cycle MEE quantum well sample 1: (b) quantum island, sample 2. The micro-PL had a spatial resolution of 1 μm.

suggests that the type A quantum well fluctuations contribute mainly to the high-energy side, while B-type islands make up the low-energy tail due to their deeper confinement potentials.

Using conventional MBE with elemental sources it seems to be impossible to suppress type A islanding, leading to rather broad QW emission lines. However, employing our method, type A island formation could be suppressed when using migration-enhanced growth. The PL of the QW shown in Fig. 2(a) has a line width of only 16 meV at an emission energy of 2.473 eV. Usually, one would expect a line width well in excess of 25 meV. This is already an indication of a very good homogeneity of the structure.

The weak localization in sample 1 is very well demonstrated by the absence of sharp discrete lines in the micro-PL shown in Fig. 2(a). Existing fluctuations have lateral dimensions of the order of the exciton diffusion length and the excitons behave like quasi-free particles, which is also seen in the temperature dependence of the PL peak shift [5].

With the help of the compositional evaluation by lattice fringe analysis (CELFA) [7], given in Fig. 3, we can obtain the Cd profile along the growth axis shown in Fig. 4. Samples 1 and 3 basically show no conventional island formation. A few island like structures were observed in sample 3, which are caused by Cd interdiffusion along dislocation lines.

Conventionally grown CdSe-based quantum structures show a strong intermixing with the ZnSe barrier material. Diffusion and segregation can lead to Cd distribution profiles with a strong tailing in growth direction [2], as shown in Fig. 3. QWs nominally consisting of up to 4 ML CdSe are broadened to 10–12 ML almost independently of the total amount of CdSe deposited, while the central Cd concentration increases with CdSe deposition.

We observe a similar behavior considering intermixing and broadening. However, Cd segregation is considerably reduced which is indicated by the sharpness of the interfaces and the symmetry of the Cd profiles which show a distinct plateau for the two samples in Fig. 4. We find an average Cd concentration of 35% and an average QW thickness of 13 ML for sample 1. Sample 3 has
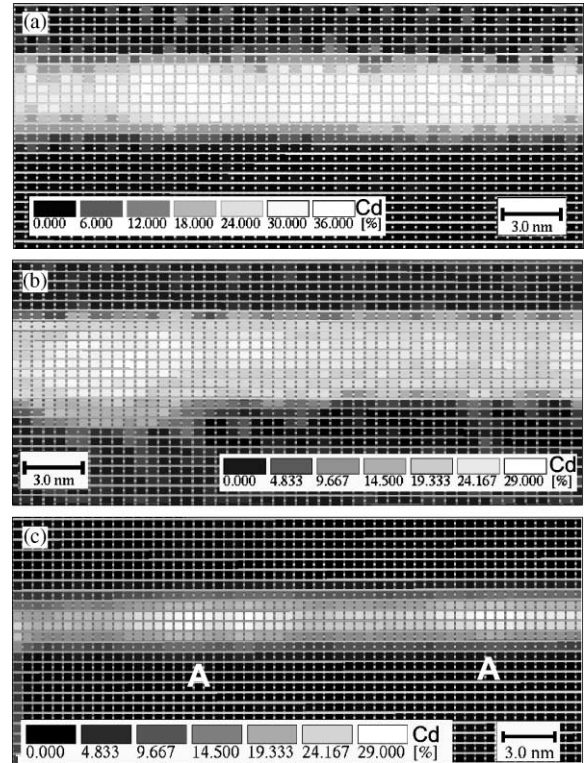

Fig. 3. Results of CELFA, the grayscale corresponds to the Cd concentration in %, the square grid marks 0.5 nm steps. (a) Sample 1, (b) Sample 3, (c) Sample 2.
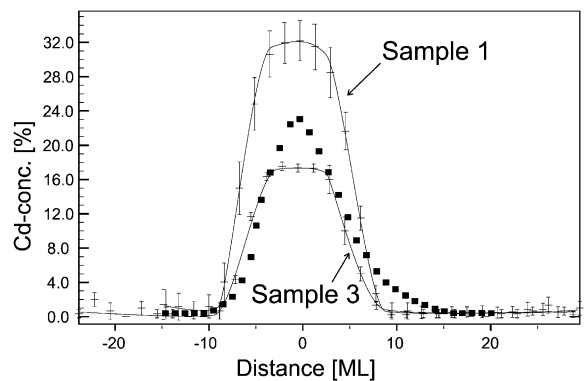

Fig. 4. Cd concentration profiles in growth direction obtained by CELFA for samples 1 and 3 according to Fig. 3. For comparison, the full squares represent data of a conventionally grown CdSe QW with 3 MLs from Peranio et al. [2].

a nominal CdSe deposition of $\frac{2}{3}$ of that of sample 1, shape and broadening are similar, but the average Cd concentration is 17%, and the average QW thickness 12.5 ML.
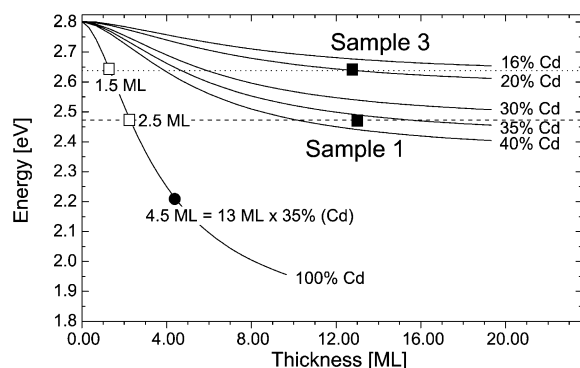
Fig. 5. Calculated heavy hole transition energies at 4 K for various Cd concentrations. Full squares represent the data (PL and CELFA) observed for samples 1 and 3, open squares mark the QW thicknesses for 100% CdSe layers of similar emission energy, and the circle corresponds to the transition energy of sample 1 if the layer consisted of pure CdSe.

Calculating the theoretical transition energies at 4 K employing an extended square potential model [8], which includes strain- and energy-dependent effective masses, and assuming a valence band offset between CdSe and ZnSe of 16%, we find a rather good agreement with the data obtained by CELFA as shown in Fig. 5. The nominal growth rates obtained by RHEED have to be revised as the number of RHEED oscillations observed does not correspond to the actually deposited number of layers and is misleading because of Cd interdiffusion during the MEE cycles. In order to suppress Cd/Zn interdiffusion, the MEE cycle periods need to be shortened as well as the capping duration should be reduced to a minimum.

4. Conclusion

In conclusion, we have demonstrated that the use of a CdS compound source instead of the conventionally used Cd elemental source opens new possibilities to grow high-quality CdSe-based quantum heterostructures. In particular, it was possible to find a growth mode where Cd segregation as well as the formation of short-range quantum well fluctuations are strongly suppressed.

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