Suppression of lateral fluctuations in CdSe-based quantum wells

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We report a reduction of inhomogeneous broadening in CdSe-related quantum wells in ZnSe by employing a growth technique that uses a CdS-compound source instead of the standard Cd elemental source for molecular-beam epitaxy. Assisted by the low sticking coefficient of sulfur and possibly an exchange reaction between S and Se, only a small S contamination is observed. A comparison with standard layers reveals an increase in quality and homogeneity by a strong reduction of the photoluminescence (PL) linewidth. Samples obtained by our method show extremely little lateral confinement as indicated by a lack of sharp single dot emission lines in micro PL and the absence of the extensive redshift observed in temperature dependent PL of fluctuating well potentials. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1394172]

Simple CdSe/ZnSe quantum wells (QWs), grown by molecular beam epitaxy (MBE) with elemental sources, show a strong tendency for Cd segregation and interdiffusion[1,2] leading to intermixed (Cd,Zn)Se layers. Furthermore, the formation of small Cd enriched islands[3] is observed even when not intentionally attempting to obtain quantum islands (QIs). These QI act as lateral, quasi-zero-dimensional localization centers at low temperatures, resulting in a strong inhomogeneously broadened photoluminescence (PL) emission and an ''S-shaped'' temperature dependence of the PL peak energy. An approach to improve such heterostructures is to completely avoid island formation and create QWs with smooth interfaces and a homogeneous Cd distribution to suppress lateral localization. In this letter we demonstrate the advantages of an alternative growth technique which introduces sulfur in the growth process by substituting the conventional Cd elemental source by a CdS-compound source during molecular beam epitaxy (MBE).

The CdS-compound source is operated at 650 °C as compared to typically 270 °C of the Cd elemental source. Atoms/molecules arriving at the growing surface thus carry a higher thermal energy. It has been shown that the sublimation of sulfide compounds results in sulfur dimers and metal atoms.[4] S_2 has a much lower sticking coefficient than Se_x ($x \geq 2$) provided by the Se source. Direct competition on the growing surface during simultaneous exposure to CdS and Se leads therefore to an incorporation of Se rather than sulfur. Furthermore, an exchange reaction can occur between on the surface physi- or chemisorbed CdS and Se molecules. Thus at the given growth temperature of 280 °C only a small amount of sulfur of less than 2% is observed in the structures.[5] The samples investigated were grown on epiready, etched and thermally deoxydized GaAs substrates. Buffer and cap layers of ZnSe were grown using elemental Zn and Se under stoichiometric growth conditions, applying a 30 min growth interrupt under Se flux prior to quantum structure growth.

We observe an overall improvement in quality of our CdSe-based heterostructures when using the sulfide. The reason is not well understood, however, a surfactant-like effect or changed energy states of the reacting atoms/molecules may be responsible. The effect on the island formation in a continuous MBE growth mode, i.e., simultaneous supply of CdS and Se, leads to results equivalent to CdSe/ZnSe based structures grown with low temperature atomic layer epitaxy (ALE)[6,7] without the necessity of applying a temperature sequence. In order to avoid island formation in thin QWs, which is partially governed by the tendency for Cd segregation, we have used migration enhanced epitaxy (MEE) with long Se exposure times (typ. 30 s–1 min) to completely chemically bind excess Cd on the surface. The samples obtained are still subject to the typical CdSe/ZnSe intermixing due to Cd/Zn interdiffusion[1,2] and a broadening of the intentionally pure CdSe layers to about 13 ML of (Cd,Zn)Se. The results by transmission electron microscopy (TEM) and chemical evaluation by lattice fringe analysis (CELFA)[8] are shown in Fig. 1. A sample overview is given in Table I. Segregation was suppressed in the MEE mode, as seen by the very symmetric Cd concentration profiles with steep flanks of samples A and B in Fig. 1. The Cd distribution is rather homogeneous and average Cd concentrations of ∼18% and ∼34% are found for samples A and B, respectively. No QIs are observed in TEM except for areas where the QW was penetrated by a stacking fault or dislocation as is seen on the left-hand side of the CELFA of sample A in Fig 1, consistent with Ref. 9.

In Fig. 2 we compare the PL properties of various samples. Samples containing small QI like fluctuations, i.e., QI sample C, grown by CdS/Se MBE, and QW samples D and E, grown by Cd/Se ALE, have strong inhomogeneously broadened PL linewidths, indicating QW fluctuations or QI formation. The introduction of sulfur in the MBE growth process of sample C has already led to a reduction (typ. of the order of 10–20 meV) of the PL linewidth as compared to standard Cd/Se-MBE samples and comparable linewidth to Cd/Se-ALE growth, indicating a more homogeneous distri-

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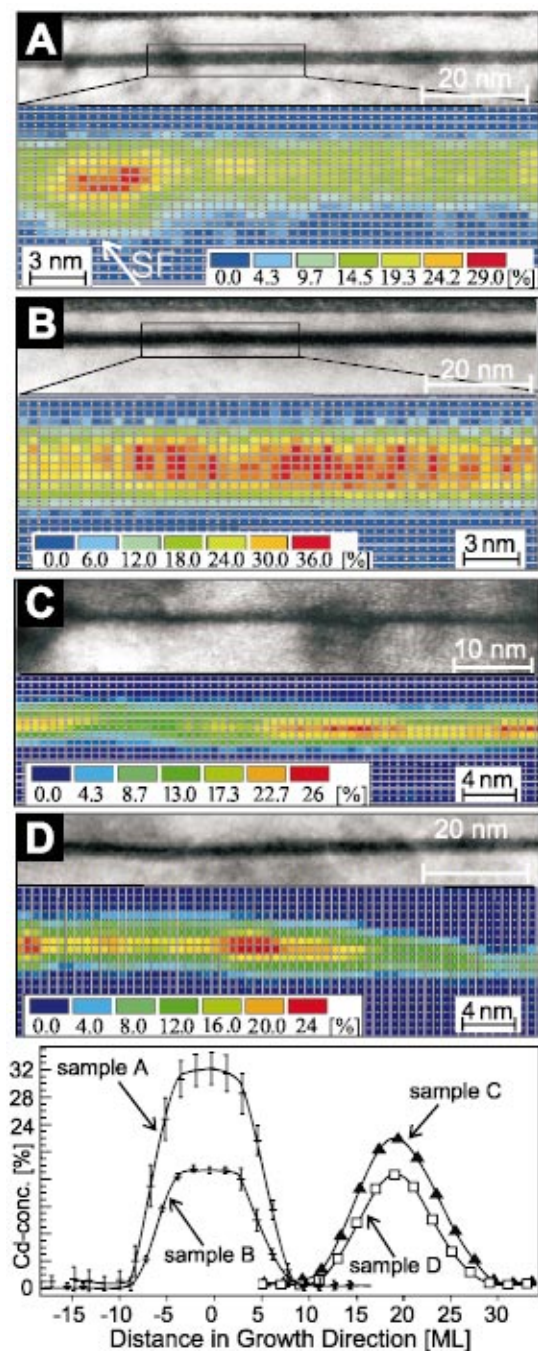[a)]Electronic mail: elisabeth.kurtz@physik.uni-karlsruhe.de

FIG. 1. (Color) (002) dark field TEM images (gray scale) and color coded Cd concentrations in % obtained by CELFA for samples A–D. Lowest: Cd concentration profiles for samples A–D. Distributions for C and D are shifted right for clarity.
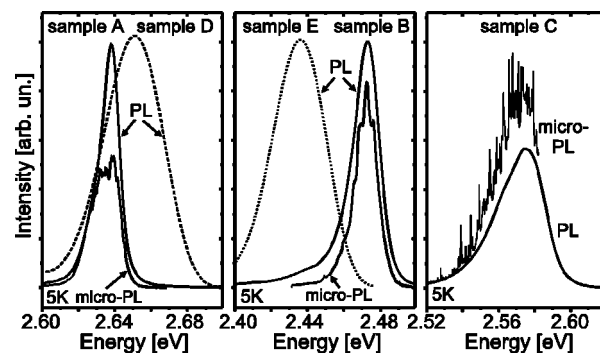

FIG. 2. PL spectra for samples A–E and micro PL for A–C (spatial resolution~1 μm, excitation at 458 nm).

bution of QI sizes. Clearly visible are resolution limited ($<200 \mu$eV) distinct lines in the micro-PL spectra of sample C which correspond to excitons localized in individual QIs. The large number of lines is consistent with the rather high density of small islands of the order of 10^{11}cm^{-2} in sample C. In case of samples A and B only few moderately sharp lines (~ 2 meV) are observed, the micro PL basically reproduces the macro PL. This indicates the absence of strongly confining lateral potentials. It can be assumed that the extension of QW fluctuations is of the order of the exciton diffusion length. Thus excitons behave like quasifree particles in the x-y plane, which is further demonstrated by the temperature dependent PL energy shift shown in Fig. 3(a). In the rough quantum well of sample E photogenerated excitons are immediately captured by the nearest potential minima at low temperatures. Thermal activation leads to an enhancement of phonon assisted relaxation[10,11] to deeper neighboring potential minima up to temperatures of 120 K and accounts for the strong redshift of the transition peak with increasing temperature, in contrast to sample B. A further temperature increase eventually leads to a thermally activated transition into higher quasicontinuous energy states above the mobility edge and the energy shift follows the behavior expected for the (Cd,Zn)Se band edge. The full width at half maximum (FWHM) of the emission peak, shown in Fig. 3(b), illustrates this evolution. A pronounced inhomogeneous broadening is observed which increases with temperature up to 150 K. The FWHM is largest for temperatures where both localized and delocalized states are populated.

In the case of the homogeneous QW of sample B lateral localization is considerably weaker and the temperature dependent peak shift directly follows the band edge shift. Only

TABLE I. Overview of all samples discussed. (Abbreviations: T_g = growth temperature; d_{nom} = nominal CdSe deposition; ΔE = FWHM of the PL peak; d_{QW} = QW thickness from CELFA; n_{Cd} = average Cd concentration from CELFA; $E_{\text{hh}}^{\text{theo}}$ = theoretical heavy hole exciton transition energies based on CELFA. Any additional lateral confinement was neglected for QI energies.)

Sample	Growth method	T_g	d_{nom}	$E_{\text{hh}}^{\text{PL}}$	ΔE	d_{QW}	n_{Cd}	$E_{\text{hh}}^{\text{theo}}$
A	CdS/Se MEE	280°	~2 ML	2.638 eV	12 meV	13 ML	18±2%	2.66 eV
B	CdS/Se MEE	280°	~3 ML	2.473 eV	16 meV	13.5 ML	34±3%	2.49 eV
C	CdS+Se MBE	300°	2 ML	2.575 eV	30 meV	11 ML	QW: 17±2%	2.69 eV
							QI: 26±2%	2.57 eV
D	Cd/Se ALE	300°	~1.5 ML	2.652 eV	32 meV	9 ML	QW: 9±2	2.76 eV
							QI: 22±4%	2.65 eV
E	Cd/Se ALE	300°	~3 ML	2.443 eV	33 meV	⋯	⋯	⋯

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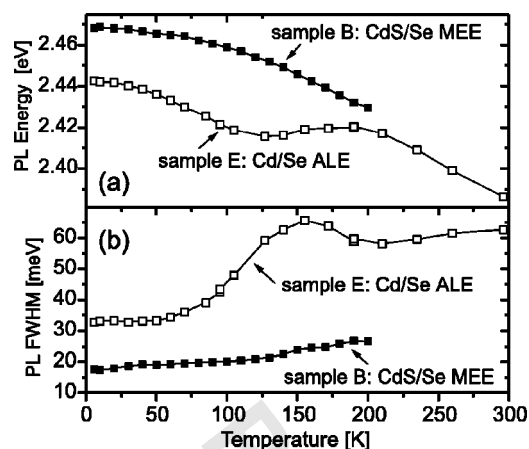

FIG. 3. Temperature dependence of (a) the peak energy and (b) the full width at half maximum (FWHM) of the PL for samples B and E.

a minor kink is observed both in the peak energy shift and the FWHM at around 40 K. An evaluation of the FWHM results in an average QW fluctuation of less than 1/10 ML. This implies, that potential fluctuations occur either on an atomic scale, where the exciton is averaging over its Bohr radius, or on a larger scale of the order of the diffusion length of the exciton corroborating the assumption that excitons can be treated as quasifree particles in the two-dimensional QW plane.

As shown in Table I, calculated transition energies for QWs according to Ref. 12 using the parameters for composition and well width deduced from CELFA measurements are well in agreement with the observed PL emission energies. If QIs are present, excitons become localized in their potentials and the transition energy observed corresponds to regions with the highest Cd concentrations. The agreement is best if the Cd concentration and distribution entering the calculation correspond to that of islands. The slight discrepancy for the QW energies obtained for samples A and B may be explained by the fact that the possibility of lateral localization was generally neglected in the determination of parameters like valence band offsets from samples grown by conventional Cd/Se MBE or ALE. The assumed valence band offset between CdSe and ZnSe of 16%[13] may therefore

be underestimated. Values in the range of 10%–30% have been suggested by various authors.[13–16] The larger value of 30% would be in better agreement with our QW results.

In conclusion, the employment of migration enhanced CdS-assisted MBE leads to CdSe:S/ZnSe quantum structures with properties superior to structures obtained by standard CdSe MBE or Cd/Se ALE. Despite the improved homogeneity of samples grown with our technique, the driving forces causing CdSe/ZnSe alloying remain an important issue.

[1] N. Peranio, A. Rosenauer, D. Gerthsen, S. V. Sorokin, I. V. Sedova, and S. V. Ivanov, Phys. Rev. B **61**, 16015 (2000).

[2] D. Litvinov, A. Rosenauer, D. Gerthsen, and N. N. Ledentsov, Phys. Rev. B **61**, 16819 (2000).

[3] D. Schikora, S. Schwedhelm, D. J. As, K. Lischka, D. Litvinov, A. Rosenauer, D. Gerthsen, M. Strassburg, A. Hoffmann, and D. Bimberg, Appl. Phys. Lett. **76**, 418 (2000).

[4] P. Goldfinger and M. Jeunehomme, Trans. Faraday Soc. **59**, 2851 (1963).

[5] E. Kurtz, M. Schmidt, M. Baldauf, S. Wachter, M. Grün, D. Litvinov, S. K. Hong, J. X. Shen, T. Yao, D. Gerthsen, H. Kalt, and C. Klingshirn, J. Cryst. Growth **214/215**, 712 (2000).

[6] M. Rabe, M. Lowisch, and F. Henneberger, J. Cryst. Growth **184/185**, 248 (1998).

[7] E. Kurtz, T. Sekiguchi, Z. Zhu, T. Yao, J. X. Shen, Y. Oka, M. Y. Shen, and T. Goto, Superlattices Microstruct. **25**, 119 (1999).

[8] A. Rosenauer and D. Gerthsen, Adv. Imaging Electron Phys. **107**, 121 (1999).

[9] T. V. Shubina, A. A. Sitnikova, V. A. Solov'ev, A. A. Toropov, I. V. Sedova, S. V. Ivanov, M. Keim, A. Waag, and G. Landwehr, J. Cryst. Growth **214/215**, 727 (2000).

[10] F. Gindele, K. Hild, W. Langbein, U. Woggon, K. Leonardi, D. Hommel, T. Kummell, G. Bacher, and A. Forchel, J. Lumin. **83-84**, 305 (1999).

[11] A. Klockikhin, A. Reznitskii, L. Tenishev, S. Permogorov, S. Ivanov, S. Sorokin, Kh. Mumanis, R. Seisyan, and C. Klingshirn, JETR Lett. **71**, 242 (2000).

[12] H. Mathieu, P. Lefebvre, and P. Christol, Phys. Rev. B **46**, 4092 (1992).

[13] S. Lankes, T. Reisinger, B. Hahn, C. Meier, M. Meier, and W. Gebhardt, J. Cryst. Growth **159**, 480 (1995).

[14] N. T. Pelekanos, J. Ding, M. Hagerott, A. V. Nurmikko, H. Luo, N. Samarth, and J. K. Furdyna, Phys. Rev. B **45**, 6037 (1992).

[15] P. M. Young, E. Runge, M. Ziegler, and H. Ehrenreich, Phys. Rev. B **49**, 7424 (1994).

[16] H. Y. A. Chung, N. Uhle, and T. Tschudi, Appl. Phys. Lett. **63**, 1378 (1993).