**Review Article**

**Arrays of Two-Dimensional Islands Formed by Submonolayer Insertions: Growth, Properties, Devices**

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Ultrathin insertions of a narrow band-gap material in wide band-gap matrices represent a challenging medium in view of aspects of growth phenomena, unique optical properties, and non-trivial approaches for structural characterization. In a very general case ultrathin submonolayer insertions may form arrays of islands due to the principally discrete nature of the growth front. If the islands are large enough, these islands may act as locally formed quantum well (QW) insertions. If, however, the islands’ size is comparable to the Bohr radius and the band-gap difference between the insert and the matrix material is large enough, quantum dots (QD) are formed. Realization of the first or the second regime depends on the surface properties of the substrate and the deposit, particularly, on the tensors of the intrinsic surface stress of both materials and on the lattice mismatch. In this work we consider in detail the case of ultrathin CdSe insertions in wide gap ZnMgSSe matrices: that the nominal thickness is chosen below the critical thickness for three-dimensional (3D) island formation. We give an overview of the experimental results available for these structures obtained by submonolayer or about-one monolayer CdSe depositions. A comparison with similar phenomena observed in conventional III–V and III–N systems is given and possible growth scenarios are discussed. We also discuss practical device applications of the structures based on ultrathin insertions for non-traditional devices. Examples of resonant waveguiding and lasing in edge geometry, of surface emitting lasers with low finesse cavities, and of broad-miniband high-frequency Esaki-Tsu anti-dot superlattices are given.

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1. Introduction

Quantum-dot (QD) heterostructures [1, 2] i.e. semiconductor structures providing quantum confinement in all three dimensions, present the ultimate limit of size quantization in solids and result in the strongest possible modification of the electronic properties as compared to quantum wells and wires. For laser applications, the use of structures with a dimensionality lower than 2 is advantageous. It results in a more pronounced modification of the density of states [3] as compared to the quantum-well (QW) case. In the case of QDs, the $\delta$-function-like density of states and the enhanced overlap of electron and hole wavefunctions in a QD may result in decreased threshold current densities, higher temperature stability of the threshold current, as well as higher material and differential gain [4]. Enhanced nonlinear effects are also expected [5]. Currently, the interest in device application of QDs is mostly related to conventional III–V materials [6] with relatively large three-dimensional (3D) QDs formed via Stanski-Krastanow or Volmer-Weber growth on the top of wetting layer or on the bare substrate, respectively. Using this approach competitive QD lasers based on GaAs emitting up to 1.3 µm have been created [7]. Very low threshold currents are realized in vertical-cavity surface-emitting laser structures [8, 9]. With regard to this progress, there are attempts to apply the QD concept to wide-gap lasers based on II–VI materials and group-III nitrides [10, 11]. The CdSe/ZnMgSSe system represents a very good choice to study the physical mechanisms of lasing in QDs based on II–VI wide-gap compounds. As it has an extended history of investigations [12], it can be considered as a model system. Moreover, despite of significant progress in nitride-based light-emitting devices, the interest towards II–VI green lasers has been revived recently by the demonstration of reduced threshold current densities in lateral-guided ZnCdSSe lasers made by N$_2^+$-molecule ion implantation [13]. By using special Li$_3$N contacts to p-type layers and rapid thermal annealing a reduction of the threshold current down to 42 A/cm$^2$ was achieved [14], which matches the best results for QW heterostructure lasers [15]. These results underline the potential of the II–VI compounds for commercial green lasers [16]. The use of QDs becomes particularly important in this case, as they suppress
nonequilibrium carrier diffusion and, thus, may increase the operating lifetime to practically acceptable numbers. In work [17], the authors speculate that the enhanced degradation stability is related to the CdSe-rich domains having a lateral size much larger than the exciton Bohr diameter in the active region.

For the fabrication of QDs several self-organized growth approaches may be applied: islands can be formed by heteroepitaxial submonolayer deposition, by spinodal decomposition of a multi-component alloy, via Stranski-Krastanow (SK) or Volmer-Weber (VW) island growth mode, by growth on faceted or step-bunched surfaces, etc. In the case of Stranski-Krastanow growth, nominally thicker deposits are applied, resulting in relatively large (>10 nm) islands [6]. The Stranski-Krastanow mode is particularly well established in the In(Ga)As–GaAs material system. A similar approach for the CdSe/ZnSe system results in the formation of islands with diameters typically larger than 30 nm, e.g. [17–22]. Such II–VI islands are often too large in comparison to the Bohr diameter in ZnSe (9 nm) to provide a significant energy separation of QD exciton quantization sublevels. Moreover, some researchers found in atomic force or scanning tunneling microscopy that these islands may be related, in some cases, to Se precipitates rather than to CdSe QDs [24]. An alternative approach to Stranski-Krastanow growth of QDs [24] has been proposed by using submonolayer (SML) narrow gap InAs insertions deposited on vicinal [26] GaAs(100) surfaces. The islands were thought to be attracted by steps in a step-flow growth mode. It was shown, however, that the InAs growth on vicinal GaAs(100) surfaces occurs via the nucleation of islands which are not necessarily attached to steps [27]. It was also shown that such islands having a height of one monolayer are of a roughly uniform size (width of about 4 nm for InAs elongated islands) [27]. The fabrication of nanostructures on GaAs(100) surfaces by submonolayer (SML) deposition has been proposed [28] and superior optical properties as compared to slightly-above monolayer insertions have been demonstrated.

At the same time, one should note that with the narrow gap III–V compounds the lateral size of the islands of about 4 nm in width and 1 monolayer in height appears to be insufficient for electron confinement in view of the fairly small electron effective mass. The electron in this case is bound mostly by the attractive Coulomb potential of the hole bound to the island and no strong lateral confinement of localized excitons may be expected. Even in this case increased exciton binding energy [29] and oscillator strength [30] have been demonstrated. The situation was shown to improve using AlGaAs barriers [31, 32], where magneto-optical studies directly demonstrated the strong squeezing of the lateral size of the exciton bound by the SML island. For typical II–VI compounds, however, an island of already very small size can be used very effectively to localize both electrons and holes (the exciton Bohr radius is only about 4.5 nm in ZnSe). The proposal for the formation of such islands in case of II–VI SML insertions, as well as the demonstration of high exciton oscillator strength and lifting of the \( \mathbf{k} \)-selection rule has been given in [33, 34]. This idea, however, was initially met with skepticism, as the mechanisms used for III–V growth were believed to be hardly possible for II–VI materials [35]. However, high-resolution transmission electron microscopy studies and studies of the influence of deposition conditions on SML luminescence confirmed the formation of nanoscale islands having a lateral size of 4–5 nm. Islands of similar size and shape demonstrating QD properties have been revealed both in cases of conventional MBE [37–39] and MOCVD [40] growth. CdSe islands having a two-
dimensional shape and a size of about 5 nm have been reported recently for MBE growth using a migration-enhanced MBE mode [41]. For the case of conventional MBE growth it was shown in [42] using transmission electron microscopy (TEM) and cross-section high-resolution TEM (HRTEM) that the density of the islands with lateral sizes of 5–8 nm increases from \(3 \times 10^{11}\) to \(7 \times 10^{11}\) cm\(^{-2}\) with increasing CdSe nominal deposit from 0.7 to 3.6 monolayers. In the case of the 3.6 ML CdSe insertion larger islands were also observed. They had sizes of \((15 \pm 2)\) and \((35 \pm 5)\) nm in two orthogonal \((110)\) directions and a density of \(5 \times 10^9\) cm\(^{-2}\). It was also concluded that there exist regions of lower Cd content separating the CdSe-rich inclusions. The estimated effective CdSe content is 2–4 times higher in the CdSe-rich regions with respect to the regions separating them. Thus, it was possible to conclude that the interdiffusion processes upon overgrowth do not result in significant smearing of the composition profile of the QDs. High compositional contrast has also been revealed in the case of MOCVD-grown samples [43].

There are numerous attempts to explain the spontaneous formation of nano-islands being uniform in size and shape. The formation can be explained by kinetic [44] and by quasi-equilibrium models [45, 46].

A completely different interpretation of the CdSe-ZnSe SML growth was given, however, by Toropov et al. [47], who proposed that uniform quasi-alloy coverage is formed for CdSe SML depositions for average thickness below 0.7 monolayer. For depositions above 0.7 monolayer the formation of large mesoscopic islands with dimensions much larger than the exciton Bohr radius is proposed. The authors drew this conclusion from the fact that the observed narrow luminescence width is not consistent with the formation of islands, having a lateral size comparable to the exciton radius and significant scatter of lateral sizes. They also admitted that the spectral position of the luminescence is close to that expected for 1 monolayer insertions. They proposed anticorrelated growth of mesoscopic islands in the case of small spacers, and drew the conclusion based on conventional plan-view TEM that the density of these islands is only about \(3 \times 10^9\) cm\(^{-2}\) and their size is 20–60 nm [10].

As all basic properties of the laser (modal gain, lasing mechanism, screening of excitons, lifting of \(k\)-selection rule) are crucially dependent on size, density and relative arrangement of islands, it is very important to understand the true nature of SML growth to ensure the formation of a high concentration of uniform islands and evaluate the impact of vertical coupling of QDs in different sheets and other growth-related effects on optical properties of SML QDs.

### 2. Self-Organization Effects in Heteroepitaxial Submonolayer and Monolayer Growth

The spontaneous formation of ordered arrays of islands has been studied theoretically and experimentally for a long time (see e.g. review in [48]). The formation of ordered (“parquet”) structures on crystal surfaces has been shown to occur when two phases with different values of intrinsic surface stress \(\tau_{ij}\) coexist on the surface [49]. Roughly speaking, the surface of the crystal is always intrinsically stressed due to the necessity to follow the lattice parameter of the bulk where the atom arrangement is different. If the values of this surface stress are different for the two phases co-existing on the crystal surface, formation of boundaries will always result in some elastic energy relaxation.
of the more stressed phase along the boundaries between the domains, making ripening of the domains energetically unfavorable. For strained above-monolayer-high 2D islands always a total energy minimum exists for a particular island size \[50\]. Indeed, the total energy of an array of islands is given by

\[
E_{\text{total}} = E_{\text{surf}} + E_{\text{bound}} + E_{\text{elast}}.
\]

Here, \(E_{\text{surf}}\) is the sum of the surface energy of islands and of the surface energy of uncovered parts of the substrate, \(E_{\text{bound}}\) is the energy of the island boundaries and \(E_{\text{elast}}\) is the elastic relaxation energy of islands due to the discontinuity of the intrinsic surface stress tensor at the island boundaries. While \(E_{\text{surf}}\) depends only on the amount of deposited material and does not depend on the island size \((L)\), \(E_{\text{bound}}\) and \(E_{\text{elast}}\) depend on the island size. Thus, one can write

\[
E_{\text{total}} \propto -\frac{\alpha}{L} \ln(\beta L) + \frac{\gamma}{L},
\]

where \(\alpha\) and \(\beta\) are coefficients accounting for the difference in the intrinsic surface stress between the two phases and \(\gamma\) describes the effect of the short-range potential due to additional dangling bonds formed at the island edges. The equilibrium size of the island is determined by the minimum of Eq. (2) which exists for any finite positive values of coefficients \(\alpha\), \(\beta\) and \(\gamma\). In contrast, for homoepitaxial SML deposition \(\alpha = 0\), and the minimum energy corresponds to an infinite island size, meaning continuous ripening of 2D islands. The delta-function-like distribution of island sizes (island size is \(L_0\)) can occur only at zero growth temperature. At finite temperature the island size distribution somewhat broadened, and another peak in the island size distribution appears near the zero island size, corresponding to the finite concentration of free adatoms and their associates on the surface. The mean size and density of the equilibrium islands decrease with increasing growth temperature \[51\]. At very high temperatures only the peak in the size distribution curve at zero island size survives and the island size dispersion becomes very pronounced.

In case of stacked arrays of 2D nano-islands one should add in Eq. (1) another term \(E_{\text{elast}}^{\text{SB}}\) which corresponds to the elastic energy of the interaction between surface islands and buried islands. Usually, this term is much less than the other three and each sheet of islands tends to form the same periodic structure which is determined by Eq. (2). If the interaction between the surface islands and the buried islands is neglected, the surface array of islands as a whole can have an arbitrary in-plane shift relative to the position of the buried array of islands. The strain field created by buried islands has the same periodicity as the array of surface islands alone. Therefore, \(E_{\text{elast}}^{\text{SB}}\) does not change the periodicity of the surface structure, but determines the position of the surface structure relative to the array of buried islands.

Generally, two possibilities can be realized for the vertical arrangement \[37\]: vertical correlation (the islands in the surface layer are placed directly above the islands in the buried layer) or vertical anticorrelation (the islands in the surface layer are placed between islands in the buried layer). To treat the system quantitatively a growth model was developed \[52\], which was based on the following main assumptions:

1. 2D islands of 1–2 monolayer height have a fixed lateral size, and the tunable parameter is the ratio of the separation between successive sheets with 2D islands and the lateral periodicity of the islands on the surface.

2. Elastic anisotropy of the cubic crystal is included.
The elastic anisotropy is known to favor ordering of nanostructures in elastically soft directions and one can expect a significant effect of the elastic anisotropy on the vertical correlation between islands.

Since cubic crystals with zinc-blende structure have a negative parameter of elastic anisotropy,

$$\xi = \frac{c_{11} - c_{12} - 2c_{44}}{c_{44}} < 0,$$

where $c_{11}$, $c_{12}$ and $c_{44}$ are moduli of elasticity of a cubic crystal in the Voigt notation, an oscillatory decay of the strain produced by buried islands is possible. It can result in the alternation of vertical correlation and anticorrelation as a function of the spacer thickness between successive sheets of islands. Figure 1 shows a calculated phase diagram for arrays of 2D islands.

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### 3. Structural Characterization of Ultrathin Insertions

The structural characterization of ultrathin insertions represents a challenge for electron microscopists due to the difficulties to reveal tiny fractions of introduced material, with
a shape and size masked by strain effects and by the averaging effects due to the finite TEM sample thickness along the direction of the electron beam. This averaging is particularly important for heavily stepped surfaces and may deteriorate shape and composition information on the QDs. In contrast, atomic-force microscopy deals with surface structures after cooling, which may be very different as compared to the growth picture.

Fig. 2. a) (110) projection HRTEM image of a CdSe/ZnSe submonolayer (SML) superlattice structure. b) Color-coded maps of the local lattice parameter along the vertical (001) direction and c) the total atom displacements with respect to the underlying ZnSe layer plane for the same area [37]
at the applied growth temperature. Particularly for II–VI systems this technique can lead to erroneous results due to the formation of Se clusters on the surface [55] and other artifacts.

We illustrate below how the structural evaluation helps to distinguish very tiny insertions of the strained material. Processed high-resolution transmission electron microscopy (HRTEM) is a powerful tool to reveal the structural properties of structures with ultrasmall QDs. In Fig. 2 we show an HRTEM image (Fig. 2a), and the same image processed using the DALI [56] technique (Laboratory of Electron Microscopy, University of Karlsruhe). The color-coded maps correspond to the averaged local lattice parameter in the growth direction (Fig. 2b) and to the total averaged atom displacements with respect to the reference lattice of the underlying ZnSe layer (Fig. 2c).

Although, the islands cannot be immediately identified in the original HRTEM images, the DALI processing allows clear visualization of QDs as regions of increased local lattice parameter. Similar conclusions can be deduced from the color-coded maps of the total atom displacement pointing to the bending of the atomic planes in the cap region caused by the strained island [37]. In the case of CdSe–ZnSe insertions, a correlated growth of islands was observed using the DALI HRTEM image processing for spacer layer thickness <3 nm, while anticorrelated growth occurred for spacers thicker than 3 nm [34, 53, 57]. The lateral size of the QDs was about 3 nm for ZnCdSe QDs in a ZnMgSSe matrix, and about 4–5 nm in the case of a ZnSe matrix. Similarly sized CdSe islands have been revealed in [41]. For MOCVD-grown QDs the lateral size varied between 2 and 6 nm and the PL emission from ultrathin (0.5–2 monolayer) insertions was broader [58]. Vertically correlated growth of MOCVD-grown islands for small spacer layer thickness has been reported in Ref. [40].

4. Optical Properties of CdSe Ultrathin Insertions in a (Zn, Mg) (S, Se) Matrix

4.1 Single-sheet submonolayer insertions. Proof of individual quantum dot

3D quantum confinement of excitons in 2D-shaped nano-islands produces an atom-like energy spectrum of excitons [59]. For very dense arrays of QDs, e.g. as formed by SML insertions, it may be difficult to resolve luminescence lines originating from individual QDs. Studies of single QD luminescence can be performed using spot-focus catho-

Fig. 3. Low temperature (20 K) cathodo-luminescence (CL) spectra of single 1 monolayer CdSe insertion in a ZnSSe matrix. The sharp luminescence lines in the spectra originate from single quantum dots (QDs) [58]
doluminescence (CL) studies. The CL spectrum of a MOCVD-grown sample with a 1 monolayer CdSe insertion in a ZnSSe matrix is depicted in Fig. 3 [58]. The FWHM of the sharp emission lines is limited by the spectral resolution. The lines originate from transitions in single QDs. Since the density of QDs and, thus, that of single sharp lines is still too high in the case of CL excitation conditions, a broad luminescence background is also detected. To resolve luminescence of only a few individual QDs, a further reduction of the investigated area is necessary. One possible way to perform such measurements is to use small etched mesas. Ultrasharp luminescence lines due to single QDs and a high density of nanoscale QDs formed by 1–2 monolayer CdSe deposition in a ZnSe matrix using MBE-growth have been observed in [41]. Measurements on mesas, however, suffered from pronounced nonradiative recombination at the mesa side-walls, particularly, at elevated temperatures. Damaged regions near the mesa side-walls may also result in an underestimation of the number of islands per mesa. To avoid nonradiative leakage of nonequilibrium carriers at the mesa side-walls while keeping the investigated area small, one can use an approach of ultrasmall openings in metal masks [60]. This technique had been used to resolve single QD emission lines up to high observation temperatures and to calculate the density of the QDs. The results appeared to be in a good agreement with the values obtained from HRTEM studies. A series of temperature dependent spectra of a single QD is displayed in Fig.4a [61]. Increasing the temperature enhances the probability of phonon-related dephasing processes, causing Lorentzian broadening of the lines above 50 K. For temperatures above 110 K the lines were still clearly resolved in the spectra while their overlap became more pronounced. The peak energy of single lines and of their overlap at higher tem-

![Fig. 4. a) Emission spectra of an individual CdSe QD for different temperatures. b) Temperature dependent linewidth of individual QD exciton lines. c) Temperature dependent integrated intensities of individual QD exciton lines [61]](image-url)
temperatures followed the CdSe band-gap dependence up to room temperature, pointing to the fact that no change in the recombination mechanism took place. A lineshape analysis showed that the integrated intensity remained almost constant (see Fig. 4c) up to and above 100 K, while the amplitude decreased due to the dephasing-induced broadening. These observations suggest that thermal activation of QD excitons to continuum states is negligible up 100 K. The FWHM of the exciton emission line as a function of temperature (Fig. 4b) was measured and modeled indicating acoustic and LO-phonon scattering to cause the broadening of the single QD lines.

4.2 Polarization of edge emission and symmetry of the heavy hole wavefunction in coupled and uncoupled quantum dots

The polarization of the photoluminescence (PL) in edge geometry enables a clear distinction between the QW and QD cases. According to Kane’s selection rule, the heavy-hole exciton luminescence in zinc-blende QWs grown on a high-symmetry (100) surface must be completely TE-polarized. This was confirmed experimentally in numerous studies. In contrast, for spherical QDs no polarization of QD emission in edge geometry can be expected. As the CdSe islands keep essentially a two-dimensional shape, the quantization in growth direction prevails, and the heavy-hole-like QD exciton luminescence in edge geometry is still TE-polarized. However, contrary to the QW case, a significant contribution of the TM-polarized emission has been observed (Fig. 5), pointing to a significant role of the lateral exciton confinement. The most remarkable observation has been done, however, for the edge emission of vertically coupled SML QDs (see Fig. 5c). This emission was found to be predominantly TM-polarized. This indicates that the heavy-hole-like exciton wavefunction was more extended in the growth direction and, most probably, had a cylindrical shape. A similar polarization of edge emission has been observed in the case of vertically coupled InGaAs–GaAs Stranski-Krastanow QDs [62]. These obser-

![Fig. 5. Linearly polarized photoluminescence (PL) of structures with 80, 30 and 15 Å spacers measured in edge geometry. The polarization changes from mostly TE for uncoupled islands (80 Å spacers) to mostly TM (accompanied by a red shift) for vertically coupled islands (15 Å spacers). The 30 Å spacer sample shows emission from both types of islands](image-url)
vations point to unique possibilities for wavefunction engineering via proper vertical alignment of QDs which may be tuned by changing the spacer layer thickness and the number of QD stacks.

4.3 Magneto-optical studies of submonolayer quantum dots. Evidence of strong lateral shrinkage of the exciton wavefunction in coupled submonolayer quantum dots

The extension of the exciton wavefunction in uncoupled and coupled QD structures was estimated from the diamagnetic behavior for magnetic fields applied parallel to the growth direction [63]. In Fig. 6 we show the PL peak energies for vertically coupled and uncoupled CdSe-ZnSe SML QDs as a function of the magnetic field. One can clearly see that the diamagnetic shift for uncoupled QDs is smaller than that of the free exciton in bulk ZnSe (dotted lines) [64]. It is, however, comparable to the diamagnetic shift of the free exciton in a 60 Å Zn$_{0.6}$Cd$_{0.4}$Se QW (dashed lines) [65] (Fig. 6, the curves for the ZnCdSe QW are vertically shifted for clarity). For the coupled QDs the PL peak energy dependence on magnetic field is very weak and the diamagnetic shift is the smallest. The lateral extensions of the exciton wavefunctions were estimated [66] from the diamagnetic shifts as ≈55 Å and less than 30 Å for uncoupled and coupled QDs, respectively. These results are in good agreement with the lateral dimensions of the 2D-shaped islands observed in cross-sectional HRTEM images [53].

Following Ref. [29], Fig. 7 shows the dependence of the exciton radius versus the QW thickness. For very narrow QWs one can use the following pseudopotential:

\[ U(z) = -\frac{\hbar^2}{m_e} \theta \delta(z) , \tag{4} \]

where \( \theta = \sqrt{2m_eE}/\hbar \), \( E \) is the energy of the electron level, \( m_e \) is the electron effective mass, \( \delta \) is a delta-function. This approximation is valid for a deep and narrow QW with the well width \( L \ll a_B \) and a band offset \( V \gg E_B \) (\( a_B \) and \( E_B \) are the Bohr radius and exciton binding

Fig. 6. Diamagnetic shift of PL peak position for vertically uncoupled (closed squares) and coupled (open squares) states of QDs. Solid lines show fitting of diamagnetic shift, dotted and dashed lines are corresponding to a free exciton in bulk ZnSe [64] and a 60 Å Zn$_{0.6}$Cd$_{0.4}$Se quantum well [65], respectively. Magnetic field is directed parallel to the growth direction.
The variational solution of the Schrödinger equation with the Coulomb potential and the pseudopotential (4) can be used to derive the exciton binding energy in an ultrathin QW [29] and, consequently, to determine the exciton radius, which is inversely proportional to it. The experimental data evaluated from Fig.6 are also plotted in Fig.7 (closed and open squares for uncoupled and coupled QDs, respectively). In the case of uncoupled states for QDs in a ZnMgSSe matrix (see Fig.7, closed circle) the estimated exciton extension in the lateral direction is much smaller compared to the expected value for an ultrathin uniform CdSe/ZnMgSSe QW, in agreement with the lateral QD size of 3 nm revealed in DALI processing for this structure [67]. For the case of CdSe QDs in a ZnSe matrix (HRTEM and DALI images are presented in Fig.2), the lateral size of the islands is larger, and the diamagnetic shift is closer to the value expected for an ultrathin QW. At the same time, vertically coupled CdSe/ZnSe QDs (lateral size of about 3 nm [57]) show a much smaller diamagnetic shift as it would be expected for the QW case. Similar values for the diamagnetic shift may be expected only for infinitely narrow CdSe QWs with infinitely high potential barriers for electrons and holes. This is clearly not the case for practical structures. Thus, the strong lateral shrinkage of the CdSe QD exciton wavefunction caused by the 2D island is directly confirmed by magneto-optical studies.

**4.4 Suppression of lateral transport of excitons in submonolayer quantum dots in ZnMgSSe matrix**

In spite of the observation of luminescence lines due to single QDs, wavefunction tunneling between neighbouring QDs may occur faster than the radiative recombination. This may result in higher probability for excitons to recombine radiatively in the neighboring larger QD, having lower exciton transition energies, as it occurs also in the case of neighboring thicker and thinner QWs. This may give in particular cases an erroneous impression of QW-like photoluminescence excitation (PLE) spectra for CdSe–ZnSe QDs with resolved ground-state heavy-hole-like exciton peak. This is very different energy in bulk ZnSe, respectively). The variational solution of the Schrödinger equation with the Coulomb potential and the pseudopotential (4) can be used to derive the exciton binding energy in an ultrathin QW [29] and, consequently, to determine the exciton radius, which is inversely proportional to it.

The experimental data evaluated from Fig.6 are also plotted in Fig.7 (closed and open squares for uncoupled and coupled QDs, respectively). In the case of uncoupled states for QDs in a ZnMgSSe matrix (see Fig.7, closed circle) the estimated exciton extension in the lateral direction is much smaller compared to the expected value for an ultrathin uniform CdSe/ZnMgSSe QW, in agreement with the lateral QD size of 3 nm revealed in DALI processing for this structure [67]. For the case of CdSe QDs in a ZnSe matrix (HRTEM and DALI images are presented in Fig.2), the lateral size of the islands is larger, and the diamagnetic shift is closer to the value expected for an ultrathin QW. At the same time, vertically coupled CdSe/ZnSe QDs (lateral size of about 3 nm [57]) show a much smaller diamagnetic shift as it would be expected for the QW case. Similar values for the diamagnetic shift may be expected only for infinitely narrow CdSe QWs with infinitely high potential barriers for electrons and holes. This is clearly not the case for practical structures. Thus, the strong lateral shrinkage of the CdSe QD exciton wavefunction caused by the 2D island is directly confirmed by magneto-optical studies.
from the case of InAs–GaAs Stranski-Krastanow QDs, where the ground state transition is not resolved in PLE spectra, while it is observed in absorption and luminescence spectra [68]. It was observed, however, that by cladding and QDs with a wider-gap ZnMgSSe matrix, one could suppress tunneling of excitons and, thus realize a true QD-like PLE spectrum [69]. Figure 8 shows PL, PLE and optical reflection (OR) spectra of the structure with CdSe/ZnMgSSe SML superlattice with relatively large spacer layer thickness at 14 K. The PL peak at 2.93 eV corresponds to the ZnMgSSe matrix as it is also confirmed by PLE and OR spectra. The OR spectrum demonstrates a clear feature at the SML superlattice ground state exciton energy (2.805 eV). The intensity maximum of the PL peak at 2.800 eV is very close to this energy. At the same time no peak is revealed in the PLE spectra in this region. The ground state which fits to the detection energy can be populated either resonantly or via the excitation in the nearest excited state that can be shifted significantly to higher energies from the ground state absorption (or OR resonance) energy.

Thus, the first peak in the PLE spectrum (2.83 eV) corresponds to the first excited state which can be also seen in the OR spectrum exhibiting modulation at this energy. We found that in the case when the exciting light and the detected emission have different polarization (\(\sigma^+\sigma^-\)) the peak at 2.83 eV is more pronounced as compared to the \(\sigma^-\sigma^+\) case. Thus this state has predominantly a light-hole-like exciton character LH0. The next excited state at 2.885 eV is more pronounced in the \(\sigma^-\sigma^+\) geometry, and it can be attributed to the excited heavy-hole-like exciton state HH1.

To prove our assumption about light- and heavy-hole character of the excited state, the linear polarization of the edge luminescence was studied (see Fig. 9). In this geometry the spontaneous emission peak corresponding to the HH0 transition (2.69 eV at 291 K) is TE-polarized, confirming its predominantly heavy-hole exciton character and the 2D shape of the nano-islands. The second peak at higher energy, which becomes dominant at high temperatures, is TM-polarized and fits to the light-hole exciton state revealed in the PLE spectra at 2.83 eV at 14 K. The lasing always occurs via the HH0 transition and is predominantly TE-polarized. A weak peak appears on the low energy tail of the spectra in the edge geometry, which is slightly TM polarized. We attribute
4.5 Lifting of \( k \)-selection rule for exciton recombination at high lattice temperatures

In bulk material and QW structures direct radiative recombination of excitons with finite \( k \)-vector, which dominates at high temperatures and high excitation densities, is forbidden. A third particle, e.g. a LO phonon, is necessary to accommodate the exciton \( k \)-vector (Fig. 10) [70]. Thus, exciton–phonon scattering processes dominate the gain mechanism in II–VI materials, where the exciton Bohr radius is small and, thus, the densities necessary to screen excitons are higher than the excitation density at the gain threshold [71]. At even higher excitation densities, exciton–exciton and exciton–electron scattering processes dominate. These processes shift the lasing wavelength typically by one

Fig. 9. Linearly polarized PL spectra at 291 K in the edge geometry below the threshold excitation density. The low energy peak corresponds to vertically coupled quantum dots

this peak to the vertically coupled CdSe–ZnSe QDs whose concentration significantly increases when the spacer layer thickness decreases. Depolarized emission in this case can be explained by the fact that the heavy-hole wavefunction becomes more symmetric for coupled states and the conventional (QW-like) selection rules for radiative recombination become inappropriate.

Fig. 10. Exciton–polariton dispersion curve illustrating the \( k \) conservation law for radiative annihilation of excitons in bulk and QW cases
or two LO-phonon energies towards longer wavelength as compared to the exciton energy revealed in the absorption spectrum. As the excitons in QDs do not have a possibility to move freely both in-plane and in vertical direction, the $k$-selection rule is not appropriate now and the gain mechanism can be different from that for bulk material. In QDs the gain mechanism has principally pure excitonic (or biexcitonic) character. This character (resonant to the QD exciton ground state) of lasing in CdSe/ZnSe QDs formed by SML deposition was first proven experimentally in [33]. As can be seen from Fig.11 in the CdSe/ZnSe SML superlattices the lasing energy is found to be very close to the exciton resonance energy in OR spectra (marked by the arrow) ($\Delta E \approx 1.4–9 \text{meV}$) even for long cavity lengths (0.8 mm).

### 4.6 Resonant waveguiding and lasing

The high exciton oscillator strength of structures with stacked dense arrays of QDs and the experimentally proven resonant character of lasing up to room temperature [69] stimulated the idea of creating a new type of a laser based on the concept of resonant waveguiding and lasing. It was shown that a significant enhancement of the refractive index originates on the low-energy side of the absorption spectrum in accordance with Kramers-Kronig relations, see e.g. [72]. In Fig.12a we show the absorption peak caused by multiple SML CdSe/ZnSe insertions evaluated from the optical reflectance spectra [73] and the corresponding $n$ modulation derived from the Kramers-Kronig equation. In the case of carrier injection, the absorption peak is partly suppressed due to the partial population of QD states with electrons and holes and a gain peak appears on its low-energy side, as larger islands provide higher capture efficiency. At finite temperatures there also exists a hopping transport of nonequilibrium carriers towards such lower energy states. In the case of excitation densities far below those corresponding to the gain saturation level, the appearance of the gain peak increases the maximum $n$ value (see Fig.12b) due to summing up positive contributions to $n$ originating on the low-energy side of the absorption peak and on the high-energy side of the gain peak. These facts enable a waveguiding in a narrow spectral window. Contrary to the case of
a conventional waveguide formed by two layers having a lower refractive index than the core layer, the enhancement of the refractive index in the active region happens in the waveguide for resonant emission processes to fit in the spectral window mentioned above. Thus, the resonant character of the lasing mechanism in QDs is an important condition for such wave-guiding. Sufficiently high absorption and/or gain values are necessary to provide significant waveguiding. Absorption measurements demonstrated maximum absorption coefficients of the order of $10^5$ cm$^{-1}$ in the case of stacked SML QDs. This fact agrees with the estimate of the exciton oscillator strength from the optical reflectance spectra which corresponds to the values of the refractive index en-

![Graph of absorption and refractive index](image1)

**Fig. 12.** Exciton-induced modulation of absorption and refractive index for the case when a) no excitation is applied and b) for the case of carrier injection. The estimated optical losses (with negative sign) due to GaAs substrate absorption are shown by a dotted line. The region where losses cross the gain curve corresponds to the expected lasing energy.

![Graph of threshold excitation density](image2)

**Fig. 13.** Dependence of the threshold excitation density for resonant waveguide laser versus laser stripe length.
hancement of about 0.2–0.3. The efficiency of the exciton-induced waveguiding is sufficient to realize lasing in both cases of II–VI, e.g. [69], and III–V [31] SML structures. Figure 13 demonstrates the dependence of the threshold excitation density ($P_{\text{th}}$) on the stripe length ($L$) at 16 K for the structure with stacked CdSe/ZnMgSSe SML QDs. As it was shown in [75] $P_{\text{th}}$ can be written as

$$P_{\text{th}} = \text{const} \left[ (\Gamma \beta J_0 + \alpha_{\text{int}}) + \frac{1}{L} \ln \left( \frac{1}{R} \right) \right],$$

where $\Gamma$ is the confinement factor of the light, $\beta$ is the gain constant, $J_0$ is the nominal current density, $\alpha_{\text{int}}$ is the internal losses and $R$ is the reflectance of the cleaved facet mirrors. $R$ is calculated from the refractive index of ZnMgSSe at the lasing wavelength and $\ln(1/R) = 0.81$ is used. From the plot of $P_{\text{th}}$ versus $L^{-1}$ shown in Fig. 13 we obtained $[\Gamma \beta J_0 + \alpha_{\text{int}}] = 18 \text{ cm}^{-1}$. Assuming that radiative and nonradiative losses are also significant, the actual internal losses in this structure must be much less than 20 cm$^{-1}$. The internal losses close to the estimated value are compatible with the internal losses in the structure with external optical confinement provided by thick wider band-gap layers with smaller refractive index [75].

4.7 Surface lasing without Bragg reflectors and self-adjustment of the cavity mode

The high exciton absorption coefficients in QDs convert to ultrahigh exciton/biexciton gain coefficients at high excitation densities, as no screening of excitons takes place. This allows to achieve lasing for very short cavity lengths in edge geometry, or enables surface lasing in vertical geometry even when no highly reflecting Bragg mirrors are used. For example ZnMgSSe/GaAs and ZnMgSSe/air interfaces could lead to 30% reflectivity. Each made it possible to achieve surface lasing in structures with 20-fold
stacked CdSe SML insertions in a ZnMgSSe matrix [57]. The surface lasing was confirmed by the excitation dependent behavior of the edge and surface emission (Fig. 14). It is important to note that the narrowing of the PL emission line and the strong superlinear growth of the intensity, both pointing to stimulated emission, have been observed first in edge geometry. PL recorded from the surface at low excitation densities shows a linear dependence of the intensity on the excitation density. Once the stimulated emission in edge geometry starts, the PL intensity in surface geometry increases sublinearly. At high excitation densities the edge emission saturates, resulting in an effective decrease of the gain [57]. Exactly at these excitation densities the surface emission starts to show superlinear growth of the intensity and a significant narrowing of the PL spectrum. Both effects, together with the saturation of the stimulated emission in edge geometry, give a clear indication of surface lasing. In this sense one should note that the above mentioned stimulated emission in edge geometry is a major obstacle to achieve low-threshold surface lasing, as it reduces the lifetime for edge emission and causes partial saturation of the gain in surface geometry. The second important reason for achieving such type of lasing is the cavity self-adjustment effect. Matching between the cavity mode and the gain spectrum is necessary for lasing. In the case of a surface-emitting laser, where the separation of cavity modes is significant, one may conclude that the lasing wavelength is completely cavity-dependent. Changes in the absorption or gain spectra of the active medium may affect somehow the exact energies of the modes, due to refractive index modulation according to Kramers-Kronig equations (so called “chirp” of the lasing wavelength). In the case under consideration, however, the modulation in gain and absorption is so dramatic, that the wavelength chirp achieves huge values, comparable to the nominal cavity mode splitting [57]. This leads to self-adjustment effects between the cavity mode and the gain spectrum. In this case the cavity mode wavelength tunes with the excitation density rise, until it reaches the spectral region where gain is able to overcome the external losses. If the differential efficiency is high, and the Fermi energy is pinned above threshold, then the shift of the mode, after the lasing condition is fulfilled, can be negligibly small. Cavity mode tunability results in the fact that the lasing wavelength follows the effective temperature dependence of ZnSe band-gap, rather than that of the refractive index.

5. Application of the Submonolayer Concept to Other Material Systems

5.1 InAs submonolayer insertions in an AlGaAs matrix

As mentioned previously [27] the deposition of SML of InAs on a GaAs(100) surface results in the formation of elongated 1 monolayer-height islands, having \( \approx 4 \text{ nm} \) width and length depending on the InAs coverage. However, this island size is insufficient for electron confinement in view of the fairly small electron effective mass in GaAs. From this point of view the use of a wider band-gap AlGaAs matrix is more promising due to the heavier electron mass in this materials compared to GaAs. It improves the exciton stability and results in the possibility to realize resonant exciton waveguiding and lasing in conventional III–V materials.

Figure 15 shows TEM plan view images of InAs SML insertions in a GaAs matrix stacked with different spacer layers. The 0.5 ML InAs insertions are embedded in a GaAs layer of 1 nm thickness. The 4 nm thick AlGaAs spacer layer composition was either 30% AlAs (a) or 60% AlAs (b). The AlGaAs barriers were used to increase the
localization energy of the QD excitons with respect to the matrix states and to tune the emission wavelength. As single islands can hardly be isolated for 20-fold stacked samples, the contrast comes mostly from bunches of vertically coupled QDs. The density of the corresponding domains revealed in TEM images increases from $7.6 \times 10^{10}$ cm$^{-2}$ for the Al$_{0.3}$Ga$_{0.7}$As matrix to about $9.1 \times 10^{10}$ cm$^{-2}$ for the Al$_{0.6}$Ga$_{0.4}$As matrix [32]. At the same time no 3D-shaped islands were revealed in cross-section TEM and HRTEM images. This observation supports the formation of QDs with 2D shapes in this material system, too. The vertical correlation of islands results in an increase of the TEM contrast, making their visualization in plan view geometry easier. On the opposite, the imaging of the islands in cross-section geometry is possible only after processing of the corresponding HRTEM images. The evaluated lateral size of a single nano-island and the indium content (normalized per island monolayer) are $5 \text{nm}$ and $90\%$, respectively [32]. These values are in a good agreement with the data obtained from magneto-optical investigations which yield an extension of the exciton wavefunction of $5 \text{nm}$ in lateral and $4 \text{nm}$ in vertical directions [76], both much smaller than the exciton radius in bulk GaAs (15 nm).

Strong exciton absorption was discovered in structures with SML insertions which led to resonant waveguiding and lasing without a conventional external waveguide [31].
This is similar to the II–VI SML QD case. Saturation of the gain (see Fig. 16) may be due to, first, reduced resonant waveguiding at higher excitation densities and, second, evolution of stimulated emission in surface geometry, as discussed in Section 4.7. Moreover, the gain peak remained narrow and the QD absorption peak does not disappear up to very high pump densities (see Fig. 16), indicating no screening of QD excitons and a very high density of QD states. This is in obvious contrast to the case of a QW or bulk GaAs, where significant broadening of the gain spectrum with increasing pump density occurs. QD gain saturation was observed only at highest excitation densities due to filling of QD states available in the range of resonant waveguiding. To estimate the role of excitonic effects, a set of InGaAs/AlGaAs structures with different active regions without external waveguide were grown on GaAs substrates [32]. Using stacked InAs SML QD insertions also resulted in a low threshold lasing in resonant waveguides structures.

5.2 Vertical-cavity surface-emitting laser based on stacked ultrathin InGaN insertions

The concept of ultrahigh gain and surface lasing was successfully reproduced in nitride materials using disc-like InN-rich QDs [77]. Multiple insertions were InGaN planes embedded in GaN matrix. The intrinsic growth mechanism results in spinodal decomposition of InGaN alloy to In-rich nano-domains having a disc-like shape and a size of about 3–6 nm [78, 79], similar to that observed in II–VI SML structures. The structure was grown by MOCVD and comprised a 2.5 μm thick GaN layer followed by an active region. This region consisted of a 25 nm thick relaxed InGaN layer with a low indium content (10% In) followed by a strain-compensated multilayer InGaN/GaN structure composed of 12 periods and having the same average In content. The multilayer struc-

![Fig. 17. PL spectra of InGaN QDs a) at different excitation densities (1) 1, (2) 0.59, (3) 0.16 MW/cm², and b) PL intensity versus excitation density [77]](image)
ture was formed by temperature cycling between 730 and 860 °C, resulting in a modulated In composition profile, as the In incorporation is strongly affected by the substrate temperature. A 0.1 μm-thick GaN cap layer was deposited on top. The PL spectra and PL intensity as a function of the excitation density at 150 K are shown in Fig. 17. The reduced differential efficiency of the stimulated emission at higher temperatures makes the spectral changes related to the increase in excitation density more evident. It can be seen from the figure that all the PL spectra are modulated by the modes of the Fabry-Perot microcavity formed by the GaN/Al2O3 interface and the GaN surface. It is clearly seen that at high excitation densities (>600 kW/cm²) one of the cavity modes starts to dominate in the PL spectra and its peak intensity grows superlinearly. Single-mode emission together with a strong increase in the slope efficiency indicates the presence of a significant feedback despite the remarkably low finesse of the cavity. This is a clear demonstration of lasing in vertical direction for structures with stacked InGaN insertions, and a first demonstration of the possibility to reach surface lasing in group-III nitrides without using high-reflectivity Bragg mirrors. According to Fig. 17a, a short wavelength shift of the cavity modes is observed with increasing excitation density. The larger shift (up to 3.2 nm) is manifested for the high-energy modes, while the low-energy modes show practically no shift. This effect can be described via Kramers-Kroning equations, which relate real and imaginary part of the dielectric susceptibility, and result in a strong modulation of the refractive index in the vicinity of the lasing energy. To explain the observed shift one needs to assume a value for the change of the refractive index in the active region of 0.4. This giant value agrees, however, with the estimated maximum gain of 10⁵ cm⁻¹ to achieve surface lasing. Quantum dots are crucial for the surface lasing approach, as they permit radiative annihilation of

![Fig. 18. a) Room-temperature PL spectra of the VCSEL structure at different excitation densities. b) Dependence of the peak PL intensity on excitation density. c) DALI image of InGaN insertions in a GaN matrix](image-url)
spatially correlated e–h pairs at any lattice temperature, provided these are trapped in QDs, and also no screening of QD excitons is possible. Without both effects, realization of giant gain coefficients necessary for lasing seems to be hardly possible.

Making a bottom AlGaN/GaN Bragg reflector [80], with maximum reflectivity exceeding 90%, one has achieved room temperature surface lasing in structures with stacked InGaN QD insertions. The current geometry of VCSELs did not imply a top DBR (the GaN/air interface played this role). Nevertheless, as it was already mentioned, ultrahigh material gain in InGaN insertions made it possible to achieve surface lasing even in case of very low finesse cavities. Room-temperature PL spectra of the VCSEL structure recorded at different excitation densities are shown in Fig. 18a. A strong increase in the slope efficiency (see Fig. 18b) and a narrowing of the PL line confirm lasing. The same conclusion can be drawn from the far-field pattern of the emission, which was found to be much narrower as compared to the spontaneous emission of the same cavity. The room-temperature threshold excitation density was 400 kW/cm². We note that the lasing emission appears on the long-wavelength side of the PL spectra. Thus, the lasing occurs via localized states caused by In-rich nano-domains formed in the InGaN insertions (see Fig. 18c). In contrast, it was shown that structures grown without the bottom DBR lase on the high energy side of the PL spectra (see Fig. 17a). To achieve higher optical gain it is necessary to overcome larger external optical losses.

5.3 Esaki-Tsu superlattices based on arrays of AlAs anti-dots in a GaAs matrix

The SML insertions may be used also in electron devices. In 1970, Esaki and Tsu [82] introduced the idea of semiconductor superlattices as a material with a new energy band structure, characterized by minibands and minigaps, and predicted a negative differential drift velocity for electrical transport along the superlattice axis being determined by the width of the lowest miniband. Negative differential velocity has been observed for a GaAs/AlAs superlattice which usually consists of a sequence of a GaAs well (several tens monolayers) and an AlAs barrier (several monolayers) [83]. In Ref. [84] a new concept of an Esaki-Tsu superlattice has been proposed and observation of miniband transport in a GaAs/AlAs superlattice with half monolayer AlAs barriers has been reported. It leads to an increasing tunneling transparency of the barriers, as compared to one or several-monolayer thick barriers, while keeping the potential barrier heights constant. Thus the expansion of the miniband width important for high-current and high-frequency applications was achieved, while the escape of electrons from the SL energy states to the 3D continuum, important for AlGaAs barriers and deteriorating the high-speed operation of the device, was avoided. At the same time no macroscopic disorder due to the AlAs lateral size and relative separation non-uniformity, degrading the overall uniformity of the structure and preventing the miniband transport, has been revealed.

In spite of the very small difference of lattice constant between GaAs and AlAs the half monolayer AlAs insertions formed on singular GaAs(100) substrates result in dense arrays of 2D-shaped nano-islands having lateral sizes of 3–6 nm — so-called anti-dots [84]. The formation of islands is not surprising as GaAs and AlAs have different intrinsic surface stress tensors even though the lattice parameters are practically coin-
ciding. The partial relaxation of the surface stress along the AlAs/GaAs domain boundary makes the formation of dense arrays of 2D-shaped AlAs nano-islands energetically favorable (see Section 2). The current–voltage characteristic (see Fig. 19) shows an Ohmic region (around zero voltage) and a region of negative differential conductivity. The behavior is characteristic for miniband transport. Record-high peak current densities were realized in the structure. These results confirm that arrays of antidots can be used for the realization of a new type of superlattices with large minibands and, thus, improved device parameters.

6. Further Prospects

The approach of using dense arrays of very small QDs achieved by submonolayer (or, more generally, ultrathin) insertions, first developed for II–VI materials, can be successfully extended to other semiconductor compounds. The latter was demonstrated for InAs SML insertions in an AlGaAs matrix and resulted in resonant waveguiding and lasing. Wavefunction engineering allowed realization of SML QDs used as active region in high power lasers emitting at 940 nm [85].

Nitrides of group III become the most attractive candidates for the application of ultrathin insertions. Such insertions have already enabled gain in the green spectral range in InGaN/GaN structures [79]. The addition of arsenic to this system [86] may help to shift the PL wavelength towards the red spectral range. In this case optoelectronic devices for the whole visible range will be possible using the same material system.

The application of the resonant waveguiding and lasing concept seems to be particularly important in systems where no convenient lattice-matched heterostructure with high refractive index contrast exists. The concept can also result in the development of very thin waveguides in the existing heteroepitaxial systems and, thus, obtaining ultralow threshold current densities.

In view of the possibility to achieve ultrahigh modal gain SML insertions are particularly promising in vertical cavity surface-emitting lasers, especially in cases when the creation of highly reflecting DBR mirrors is difficult due to a low contrast in the refractive indices of the materials used. This is the case of GaN-based VCSELs and of 1.3 and 1.55 μm emitting structures on InP substrates.

Fig. 19. Current–voltage characteristic as measured (solid line) and calculated (dotted line) for a GaAs/AlAs SML superlattice. Upper inset: principle of the measurement; lower inset: minibands calculated for a superlattice with submonolayer AlAs barriers [84]
7. Conclusion

We have discussed optical properties of ultrathin CdSe insertions in a ZnSe matrix and have transferred some major ideas to different materials systems. We have demonstrated that for appropriate growth conditions SML insertions represent dense arrays of uniform two-dimensional nano-islands with a lateral size comparable or much smaller than the exciton Bohr radius. In spite of the relatively small size of the islands, the QD characteristics are proven by direct observation of luminescence lines from single QDs up to high observation temperatures, by the resonant character of the gain and by lateral squeezing of excitons revealed in magneto-optical studies. New effects related to the ultrahigh density of SML QDs led to unique devices such as resonantly waveguiding lasers and cavity-self-adjusted surface-emitting lasers without using highly reflecting mirrors.

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