I. INTRODUCTION

Semiconductor heterostructures of reduced dimensionality like quantum wires (QW’s), and particularly, quantum dots (QD’s) have recently attracted much attention. The most promising way to fabricate QW’s and QD’s is based on self-organized growth phenomena at crystal surfaces, or in the bulk, such as periodic surface faceting, growth on faceted surfaces, spontaneous phase separation of alloys, and formation of two-dimensional (2D) and three-dimensional (3D) islands in heteroepitaxial growth. In order to display true 3D confinement effects the size of nanostructures must exceed a critical value to show confined states. On the other hand the structures must be small enough to ensure a sufficiently large energy separation of the charge-carrier sublevels to avoid thermal population of too many substates. In wide-gap II–VI materials, a small size of the islands is indispensable to obtain 2D confinement due to the large electron and hole masses. The possibilities to control the electronic states and optical properties of nano-objects by self-organization phenomena is usually, however, restricted due to a limited variation of geometrical size, shape, and degree of homogeneity. We propose an approach to overcome these limitations by using the possibility of anticorrelated or correlated growth of 2D islands.

Submonolayer (SML) insertions like CdSe in ZnSe are known to form dense and ordered arrays of nanoscale two-dimensional islands, where two-dimensional islands are considered as objects with a high width/height ratio. Formation of ordered 2D surface structures (‘parquet’ structures) caused by the coexistence of two phases on the surface, was first considered in Ref. 30. Marchenko showed that two-dimensional islands (or, e.g., domains of identical surface reconstruction), differing in the value of intrinsic surface stress from adjacent areas, always exhibit partial relaxation of this surface stress at the domain boundaries. The formation of boundaries becomes energetically beneficial. The total energy per unit surface for an array of stripes on the surface can be written as

$$E_{\text{surf}} = -\frac{\alpha}{L} \ln(\beta L) + \frac{\gamma}{L}$$

where \(\alpha, \beta, \gamma\) are coefficients accounting for the difference in intrinsic surface stress between two phases and the role of the short-range potential due to additional dangling bonds formed at the island edges. The equilibrium size of the island (\(L_0\)) is determined by the minimum of Eq. (1) which exists at any finite positive values of coefficients \(\alpha, \beta, \gamma\). In contrast, for homoepitaxial submonolayer deposition \(\alpha = 0\), and the minimum energy corresponds to an infinite island size, which means the continuous ripening of 2D islands. Formation of ordered islands using strained submonolayer depositions has been demonstrated in a number of papers for III–V and II–VI material systems. In our case the islands are strained by 7% lattice mismatch between CdSe and ZnSe. For strained covered 2D islands, partial relaxation of the elastic strain is possible near the island edges.
at the expense of some strain in the host material. Thus there exist two sources of elastic deformation of the host material, and the situation is different from the case of 3D islands where the strain around an island gradually decreases with respect to its center. This more complex character of the strain distribution around 2D islands makes it possible that the islands in the subsequent layer are placed directly above the islands in the lower layer or anticoordinated (the islands in the subsequent layer are placed between islands in the lower layer) growth.\textsuperscript{29,32,33} We note that for 3D islands, the maximum distortion of the lattice constant that favors nucleation of the island in the subsequent layer always occurs on top of the underlying 3D islands. Shchukin \textit{et al.},\textsuperscript{13} showed that the possibility of vertical correlation or anticorrelation depends on both the elastic properties of the materials used and the ratio between the in-plane and vertical separation of islands. Transition from anticoordinated to correlated growth has been predicted for spacer layer thicknesses sufficiently small with respect to the lateral period.

In this work we investigate CdSe/ZnSe SML superlattices (SL’s) with different ZnSe spacer layer thicknesses between CdSe sheets. For particular geometries of the structure we realize ordered arrays of two-dimensional islands grown either in a vertically correlated or anticoordinated way. For a narrow range of the spacer layer thicknesses a mixed situation occurs. Vertically coupled quantum islands demonstrate electronic properties which can be tuned by selecting the particular geometry of the structure.

\section*{II. EXPERIMENTAL DETAILS}

\subsection*{A. Sample growth}

The structures studied in this work are grown on $n^+$ GaAs(100) substrates using molecular beam epitaxy at a substrate temperature of 280 °C.\textsuperscript{34,35} and were partially described elsewhere.\textsuperscript{29} All structures consist of 360-nm ZnS$_x$Se$_{1-x}$ buffer and 60-nm ZnS$_x$Se$_{1-x}$ cap layers lattice matched to the GaAs substrate. Between these layers a CdSe/ZnSe SML SL is inserted. ZnSe barriers separate the sheets of CdSe SML insertions, and have thicknesses of 15, 30, 50, and 80 Å. The average thickness of a single CdSe insertion is about 0.7 ML, as evaluated by the DALI procedure. The total thickness of the SML SL region is 60 nm for structures with 30-, 50-, and 80-Å barriers, and 30 nm for the structure with 15-Å barriers. Consequently, the numbers of SL periods is matched to the GaAs substrate. Between these layers a CdSe/ GaAs~$0.06$ ZnS~$0.94$ cladding layers is negligible, and the possibility of vertical correlation or anticorrelation depends on both the elastic index change between the active SML SL and the passive layers. The estimated height of the Cd-rich insertions is about 2 and 20 nm, and may therefore exceed the lateral island size. Effect along the thickness of the foil which ranges between 5 and 20 nm, and may therefore exceed the lateral island size. The estimated height of the Cd-rich insertions is about 2 and 20 nm, and may therefore exceed the lateral island size. This more complex character of the strain distribution around 2D islands makes it possible that the islands in the subsequent layer are placed directly above the islands in the lower layer or anticoordinated (the islands in the subsequent layer are placed between islands in the lower layer) growth.\textsuperscript{29,32,33} We note that for 3D islands, the maximum distortion of the lattice constant that favors nucleation of the island in the subsequent layer always occurs on top of the underlying 3D islands. Shchukin \textit{et al.},\textsuperscript{13} showed that the possibility of vertical correlation or anticorrelation depends on both the elastic properties of the materials used and the ratio between the in-plane and vertical separation of islands. Transition from anticoordinated to correlated growth has been predicted for spacer layer thicknesses sufficiently small with respect to the lateral period.

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\section*{III. EXPERIMENTAL RESULTS AND DISCUSSION}

\subsection*{A. Structural characterization}

To investigate the structural properties of the samples we perform HRTEM measurements. A DALI processing of the lattice images is necessary to reveal the shape and size of quantum islands, as reported previously.\textsuperscript{27} In Fig. 1 we show a color-coded map of the local lattice parameter in the growth direction ($a_\perp$) for the structures with 30-Å (a) and 15-Å (b) spacer layers thicknesses. The local lattice parameters are measured for each projected unit cell with the dimensions of $a_{\parallel} \times a_{\parallel}$ - $a_{\perp}$. A reference lattice parameter is determined by which die LLP’s are normalized. The reference lattice parameter for Fig. 1(a) was chosen to be the ZnS$_x$Se$_{1-x}$ lattice constant resulting in LLP’s between 1 and 1.07. An averaged lattice parameter was determined from the whole image [Fig. 1(b)]. Therefore, normalized LLP’s smaller than 1 are observed in the ZnSe spacers. The shift in the color from blue to red corresponds to an increase of the lattice parameter in the vertical direction $a_\perp$. Thus the green, yellow, and red areas indicate (Cd, Zn)Se layers with larger LLP and, consequently, with larger Cd content, due to the larger bulk lattice parameter of CdSe (6.081 Å) with respect to that of the ZnSe (5.6697 Å). One can see that the CdSe insertions demonstrate different behaviors for these two cases. The areas with increased Cd content have lateral sizes of about $\sim$40–50 Å for 30-Å spacers and of $\sim$25–30 Å for 15-Å spacers. It should be noted that the measured Cd composition of the islands may be reduced due to the averaging effect along the thickness of the foil which ranges between 5 and 20 nm, and may therefore exceed the lateral island size. The estimated height of the Cd-rich insertions is about 2 and 4 ML. A broadening of the Cd distribution along the growth direction may be induced by steps along the electron beam or by the segregation of cadmium.

Formation of nanoscale islands by submonolayer deposition has already been demonstrated in this material
The remarkable change in the lateral arrangement of islands with decrease of the spacer layer thickness is reported here. A close inspection of Fig. 1 reveals that the islands in the upper sheets which are separated by 30-Å spacers tend to nucleate at positions between the islands of the previous sheet (anticorrelated growth) while a vertical arrangement of islands over a period of 2–5 layers is locally observed for the structure with the 15-Å spacers [Fig. 1(b)]. As recently calculated by Shchukin et al., the character of vertical ordering of two-dimensional islands depends crucially on the relative thickness of the spacer layer with respect to the lateral period of the structures. For small spacers,
a transition from anticorrelated to correlated growth occurs if the spacer layer thickness is decreased to about one-third of the lateral period. Since the lateral period for 30-Å spacer structures can be roughly estimated to be 10 nm; the transition is expected to occur at a spacer layer thickness of 3 nm. This is in very good agreement with our experimental data. We note, however, that we do not see continuous vertical chains of QD’s through the entire structure, being typical for vertically coupled three-dimensional quantum dots. Three to four periods of stacking predominantly occur. Moreover, some of the chains are tilted. The average lateral size of the islands is smaller than in the case of anticorrelated growth. We attribute these effects to the high complexity of the system. Theoretical calculations of vertical ordering of islands in Ref. 13 do not consider the influences of the strain fields produced by underlying islands on the island lateral size in the upper row, and refer only to the relative arrangement of islands. The effect of the strain on the island size can be important exactly in the case of ultrathin spacers. Nevertheless, the stacking of three periods already has a strong impact on the PL properties, as was shown for similar structures grown by metal-organic vapor phase epitaxy.

B. Electronic properties

The properties of electronic states in quantum islands are studied by optical investigations. PL spectra of structures having different spacer layer thicknesses are shown in Fig. 2. For large spacer layer thickness only one line appears. The transition energies correspond to heavy-hole-like excitons which are localized at two-dimensional nanoscale islands with lateral extensions comparable to the exciton diameter. On the other hand, at a spacer layer thickness of 30 Å, a second lower-energy luminescence line evolves. The arrangement of islands in a vertical correlation results in an efficient coupling of electronic states between vertically neighboring islands, because the main part of the wave function is extended into the barrier. The appearance of the low-energy line can thus be assigned to such chains of coupled quantum dots. The low-energy shift of the luminescence is similar to the case of vertically coupled QD’s, as demonstrated first for 3D InAs-GaAs QD’s. A further decrease of the spacer thickness results in an increased intensity of the PL due to enhanced coupling of QD’s, and in a complete suppression of the luminescence from uncoupled islands.

The optical spectra of the SML SL with 30-Å spacer shown in Fig. 3 give further information about the electronic states. The main luminescence line originates from coupled QD’s, while the main feature in the OR spectrum appears at 2.71 eV, and corresponds to uncoupled states. This can be explained by the fact that, even if the density of uncoupled QD’s is still larger than that of the coupled QD’s, an efficient hopping and tunneling of carriers from higher-lying uncoupled states results in predominant population of the coupled states. The higher density of uncoupled QD’s is confirmed by the fact that the PLE spectra of both peaks demonstrate essentially the same shape with main resonance, corresponding to heavy- and light-hole like excitons in uncoupled QD’s.

We note that in the PLE spectra of uncoupled states one can clearly see a ground state and two excited states. As shown previously by circularly and linearly polarized PL and PLE measurements, the lowest, first, and second excited states have heavy-, light-, and heavy-hole-like characters, respectively. The appearance of the second heavy-hole-like excited state HH1 can be explained by only lateral quantization, because a second heavy-hole-like subband is not possible in the case of ultrathin CdSe quantum wells. Similar spectra are observed for all structures with anticorrelated or noncorrelated growth, i.e., samples with 30-, 50-, and 80-Å spacers. The edge emission of anticorrelated or uncorrelated SML QD’s is predominantly TE polarized, because the 2D shape of the islands results in much stronger heavy-hole k quanti-
The principal advantage of SML SL’s composed of dense arrays of two-dimensional QD’s is the possibility to realize an intrinsic resonant waveguiding effect (excitonic waveguide) even without thick cladding layers having a lower refractive index. The effect is based on a resonant enhancement of the refractive index in the vicinity of the exciton resonance in accordance with Kramers-Kronig equations. Since the studied structures are grown with thin buffer layers directly on the strongly absorbing GaAs substrate, the resonant modulation of the refractive index must be remarkably strong to provide resonant waveguiding. This is possible because of the ultrahigh material gain in QD’s and their remarkably high density for stacked SML depositions. Another strict requirement to enable excitonic waveguiding is the lifting of the in-plane \( k \)-selection rule, which prohibits zero-phonon emission of excitons with finite kinetic energy that dominate in bulk and quantum-well structures at high temperatures or excitation densities. In the latter structures, phonon-assisted gain is possible\(^{37} \) but it occurs out of the energy range of efficient resonant waveguiding. On the other hand, in-plane \( k \)-selection rules are not appropriate in QD’s, and zero-phonon gain resonant to the resonant waveguiding region is possible without using of thick cladding layers with low refractive index.

Therefore, we are able to realize gain at high excitation density and observe stimulated emission in all the structures under investigation. Gain spectra with both TE and TM emission are observed for the structure with 50-Å spacers, where only the emission from uncoupled states appears. Stimulated emission demonstrates a stronger degree of TE polarization, as can be expected in view of the stronger amplification of the TE mode. Surprisingly, we observed an appearance of the TM mode peak exactly at the energy which corresponds to the heavy-hole-like exciton state revealed in the PLE and OR spectra (Fig. 5). The observation of the polarization reversal within the contour of the same stimulated emission peak is very unusual for any kinds of laser structures studied up to now. Probable explanation is the development of the collective photonic states in QD ensembles\(^{35} \), resulting from electromagnetic interaction of QD’s. This effect is predicted to cause a splitting of the gain peak into TE and TM components in arrays of QD’s having an anisotropic QD shape and (or) relative arrangement.

A comparison of the surface PL at high excitation densities and stimulated emission recorded from the edge of the structure is shown in Fig. 6. The transition energies from the QD ground state revealed in the OR spectra of the structures are shown by solid segments. The features observed in the surface emission of the 80- and 50-Å spacer structures will be discussed below. The stimulated emission originates from uncoupled QD structures with larger spacer layer thicknesses. Emission from coupled states dominates only in case
of the 15-Å spacers, where the density of the coupled states is high. The zero-phonon nature of the QD emission is confirmed by the energy positions of the stimulated emission and the gain maximum. The redshift of the stimulated emission is distinctly smaller than the LO-phonon energy, proving the zero-phonon gain mechanism for all the structures. The energy position of the gain maximum does not shift with the excitation density, manifesting an exciton gain mechanism in the exciton waveguiding region.

Gain spectra of the structure with uncoupled QD’s at excitation densities well above threshold of stimulated emission are depicted in Fig. 7. One can clearly see that the absorption peak which appears in the region of excitonic waveguiding remains even at excitation densities as high as two orders of magnitude above the threshold. Thus there still exists a spectral range of strong excitonic absorption providing an increased refractive index on its low-energy side, and, consequently, an efficient exciton-induced waveguiding. At the highest excitation densities, however, the situation starts to change. The gain saturates with increasing excitation density, and even a decrease in gain is observed. The effect is accompanied by a saturation of the edge emission. This effect can be explained by a partial saturation of the excitonic waveguiding effect. Since stimulated emission can occur only in the narrow spectral energy range due to exciton-induced refractive index enhancement, not each QD can contribute to the gain. The number of QD’s having a proper transition energy is finite, and, after filling of these QD’s, gain saturates. At very high excitation levels when all the QD’s are filled, there is no excitonic absorption and, conse-

FIG. 5. PL and PLE spectra (a) and linearly polarized spectra of stimulated emission (b) for the structure with 50-Å spacers ($T_{\text{reg}} = 7$ K, $P_{\text{exc}} = 1$ MW/cm$^2$, and $E_{\text{exc}} = 2.88$ eV).

FIG. 6. Edge and surface PL of structures with different spacer layer thickness ($T_{\text{reg}} = 7$ K, $P_{\text{exc}} = 1$ MW/cm$^2$, and $E_{\text{exc}} = 2.88$ eV). The PL energies of the low-energy transition of the 15- and 30-Å spacer samples differ due to different lateral island sizes.

FIG. 7. Gain (solid line) and stimulated emission (dotted line) spectra for the structure with 80-Å spacers ($T_{\text{reg}} = 7$ K, $P_{\text{exc}} = 1$ MW/cm$^2$, and $E_{\text{exc}} = 2.88$ eV). The inset shows the dependence of the maximum gain on excitation density.
sequently, also non-exciton-induced waveguiding on the low-energy side of the excitonic resonance. This results in a decrease of the gain.

Simultaneously with the saturation of the edge emission a new line appears in the surface PL spectra shifted by about 20 meV to the low-energy side from the exciton resonance energy (Fig. 8; see also Fig. 6). The superlinear growth of the intensity of this line occurs at already very high excitation densities. Both effects taken together hint at the observation of stimulated emission in surface geometry. This process is possible if the gain in the structure overcomes losses, which are external losses in this case in view of the small cavity thickness. These losses can be roughly evaluated as

\[ \alpha_{\text{ext}} = \frac{1}{2L} \ln \left( \frac{1}{R_1 R_2} \right), \]  

where \( R_1 \) and \( R_2 \) are the reflectivity of the surface and the GaAs-ZnSe interface, respectively. Equation (2) gives an estimate of the losses of about \( 4 \times 10^4 \) cm\(^{-1}\). Assuming that the active gain region takes only 13% of the total vertical cavity length, the modal gain should exceed \( 3 \times 10^5 \) cm\(^{-1}\). This value is huge, but can be expected for dense arrays of small QD’s having relatively small inhomogeneous broadening. In a similar structure it was shown by Aliev et al. that the optical density of CdSe SML, SL is about 0.5. Taking into account a SL thickness of 600 Å, the absorption coefficient can be estimated to be \( 1 \times 10^5 \) cm\(^{-1}\). Thus the gain can attain this value or even exceed it up to a factor of 2 due to the biexciton contribution. In this case, the modulation of refractive index due to the Kramers-Kronig equations becomes comparable with the refractive index in the surrounding matrix. This can significantly increase the reflectivity of the mirrors. Another point is the fitting of the lasing energy to the cavity mode, which must depend on the cavity thickness. For a variable refractive index within the contour of the gain spectrum, however, a self-adjustment effect takes place, and the standing-wave condition is automatically fulfilled, if the difference of the gain wavelength and the cavity mode is not too large.

The proof of the self-adjustment effect is the shift of the lasing mode with the gain spectrum. This can be realized by changing the temperature. To monitor the behavior in a wide spectral range we used a structure with CdSe SML insertions in a ZnMgSe\(_{1-x}\) matrix, providing better localization of carriers and permitting lasing up to room temperature. In Fig. 9 we show the temperature dependence of the surface lasing energy. PL spectra of the structure below and above threshold are shown in the inset. As it follows from Fig. 9, the lasing energy follows the matrix band-gap temperature dependence which is much stronger than the usual temperature dependence of the cavity mode. The latter is weak because it originates from a change of the refractive index of the matrix material. Thus the self-adjustment effect seems to be the most probable explanation of the stimulated emission in surface geometry.

**IV. CONCLUSIONS**

To conclude, we demonstrated that by proper selection of the structure geometry by using submonolayer depositions one can tune the vertical arrangement of submonolayer islands and consequently the electronic spectrum of quantum
dots. We demonstrated that the polarization of the luminescence in edge geometry is opposite for coupled and uncoupled two-dimensional quantum dots. We observed resonant waveguiding and stimulated emission in edge geometry for both types of quantum dots. At very high excitation densities we found strong evidence for stimulated emission in surface geometry in structures without Bragg reflectors, and attributed the effect to ultrahigh material gain in submonolayer quantum dots and a self-adjustment effect of the gain spectrum and the cavity mode.

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