Semiconductor quantum dots (QDs) are promising candidates for a number of optoelectronic applications. Since these applications frequently require a sufficient overlap between the light field and the gain medium, stacked QD arrays are created and employed, e.g., in laser structures. Obvi-ously the spacing between the QD planes in such an array is an important parameter defining the interaction between confined excitons. For GaAs spacers thicker than 6 nm the situation is controlled by the carrier transfer between the stacked double InAs QDs, whereas for thinner GaAs spacers the coupling between neighboring InAs QDs becomes strong enough to substantially modify the electronic structure within the QDs, and it becomes valid to call such a structure an array of quantum dot molecules (QDMs).

In this letter we present the results of a transmission electron microscopy (TEM) and photoluminescence (PL) spectroscopy study of samples with two different GaAs spacers in between InAs QD layers grown by molecular-beam epitaxy. The samples are grown by solid-source molecular-beam epitaxy on a GaAs substrate in between InAs QD layers grown by molecular-beam epitaxy. Structural and optical characterization is done by means of transmission electron microscopy and photoluminescence, respectively. Photoluminescence spectra consist at least of three well-separated optical transitions that are assigned to molecular energy terms and a substantial exciton lifetime increase is observed. Detailed spectral analysis of the transient luminescence behavior indicates “intraterritor” transitions that could be favorably used for the creation of midinfrared light sources. © 2004 American Institute of Physics. [DOI: 10.1063/1.1769077]

Coupled pairs of InAs quantum dots are grown by molecular-beam epitaxy. Structural and optical characterization is done by means of transmission electron microscopy and photoluminescence, respectively. Photoluminescence spectra consist at least of three well-separated optical transitions that are assigned to molecular energy terms and a substantial exciton lifetime increase is observed. Detailed spectral analysis of the transient luminescence behavior indicates “intraterritor” transitions that could be favorably used for the creation of midinfrared light sources. © 2004 American Institute of Physics. [DOI: 10.1063/1.1769077]
Figure 1. (a) Cross-section (200) dark-field TEM image of the reference QD sample with a 10 nm spacer thickness. (b) Cross-section (200) dark-field TEM image of a InAs QDM sample with a 5 nm GaAs spacer. The striped structure in the upper part of the figures shows the top GaAs/AlGaAs superlattice. (c) High-resolution dark-field TEM image of a single QDM after Fourier filtering. The image is formed by interference of four (200) chemically sensitive reflections to reinforce contrast from indium compositional variations. (d) Indium concentration obtained by analysis of the high-resolution TEM image along the growth direction. The ordinate is also effective for (c).

Figure 2(a) shows normalized cw PL spectra at $T = 10$ K of the QDM sample for increasing excitation densities. Figure 2(b) gives the spectral positions of the lines as obtained from multi-Gaussian fits. The labeling of the lines in Fig. 2(b) is chosen according to the expected “molecular” electronic transitions, cf. also Ref. 6. The + sign stands for the bonding (symmetric) states. Figure 2 also involves a schematic diagram of the transitions. The dashed ellipses enclosing electron and hole levels point out to the excitonic character of the PL. For low excitation densities we find a 45 meV separation between the $s_+$ and $p_+$ lines and 24 meV spacing between $p_+$ and $d_+$. Plots of the line intensities versus excitation show an almost linear dependence with saturation thresholds at 50 and 130 W cm$^{-2}$, for the $s_+$ and $p_+$ lines, respectively. The ratio of 1:3 for their integrated intensities at the threshold points reflects the relative occupation probability of $s_+$ and $p_+$ states. For the $d_+$ line the saturation is not reached within the steady-state experiment. Arrhenius plots of the temperature-dependent PL signals provide thermal activation energies of 124, 82, and 60 meV for the $s_+$, $p_+$, and $d_+$ lines, respectively. The separation between the two lines of the reference sample amounts to 35 meV and remains almost constant in the steady-state experiment, cf. Fig. 2(c).

Figure 3 illustrate the transient recombination behavior of the QDM sample. In Fig. 3(a) PL transients at the $s_+$, $p_+$, and $d_+$ spectral line positions are presented. The recombination from the $s_+$ and $p_+$ states is characterized by a monoexponential PL decay with time constants of 1.3 and 1.0 ns, respectively. The $d_+$ band exhibits a biexponential PL decay, namely a fast (0.8 ns) and a slow (1.7 ns) transient. For growing excitation densities the slow component approaches the behavior of the fast component, see Fig. 3(b). The reference sample, see Fig. 3(c), shows monoeponential PL decays with time constants of 0.55 and 0.75 ns, arising from the smaller (first layer) and larger (second layer) insulated QDs, respectively.

The results given in Fig. 1 clearly show that, at least from the structural point of view, QDMs consisting of almost equally sized QDs are achieved. According to theoretical...
ical predictions\textsuperscript{5–7} the spacer thickness of about 5 nm should lead to electronic properties corresponding to QDMs, whereas a 10 nm spacer keeps the properties insulated QDs. Low-density PL spectra from both samples show a PL doublet with 45 and 35 meV energy spacing. In agreement with our TEM data for the reference sample this doublet is likely to be caused by different island sizes in the first and second QD planes.

For the QDM sample, the 45 meV split is assigned to the energy between the \( s_\downarrow \) and \( p_\uparrow \) states. This value matches the difference of thermal activation energies between the \( s_\downarrow \) and \( p_\uparrow \) lines (124–82=42 meV). This is very similar to literature data, namely 42, 38, and 23±7 meV reported for QDM samples with about 5 nm thick spacer by Bayer et al.\textsuperscript{3} Hinzner et al.\textsuperscript{5} and Korkusinski et al.\textsuperscript{7} respectively. The observed \( p_\uparrow - d_\downarrow \) split of 24 and 22 meV obtained from PL spectra and Arrhenius analysis, respectively, indicates that \( s_\downarrow, p_\uparrow, \) and \( d_\downarrow \) transitions involve the same hole level being energetically separated from the GaAs barrier by about 190 meV (see also the scheme in Fig. 2). This value well corresponds to theoretical predictions indicating strong hole localization in QDMs.\textsuperscript{8}

As demonstrated by Fig. 1(b) the \( s_\downarrow - p_\uparrow \), and even more, the \( p_\uparrow - d_\downarrow \) splits increase for higher excitation densities. This effect is explained by a sequential filling of the \( p_\uparrow \) and \( d_\downarrow \) states with a maximum occupation of 6 and 10, respectively. The low-energy shift of the \( s_\downarrow \) line with an increasing density is likely to be caused by bie exciton formation, cf. Ref. 6.

Our tentative assignments of the PL contributions are supported by the transient PL data, see Fig. 3. We start with addressing the absolute PL decay times of 0.8–1.7 ns for the QDMs, and of 0.55–0.75 ns for the insulated QDs. The increased excitonic lifetime in the QDMs is assigned to the stronger hole localization resulting in a smaller overlap between electron and hole states, and eventually reduced oscillator strength of transitions in QDMs compared with insulated QDs, cf. Ref. 8.

For analysis of the transient PL data we take into account the density of QDMs \((1.0±0.2) \times 10^{11} \text{ cm}^{-2}\), the state multiplicity (2 for \( s_\downarrow \)), and the ratio of the PL decay times at the spectral positions of the \( s_\downarrow \) and \( p_\uparrow \) lines that amount to 1.3 and 1.0 ns, cf. Fig. 3(a). Even at \( 2.4 \times 10^{11} \) photons/pulse \((\text{cm}^2 \times \text{ns}) \) the \( s_\downarrow \) state is filled and the transition \( p_\uparrow \rightarrow s_\downarrow \), which is allowed by the selection rule \((\Delta m=1)\), is blocked. Bryllert et al. have deduced a Coulomb blockade effect from temperature-dependent current-voltage measurements at a single QDM.\textsuperscript{9} In our case, the radiative recombination of electron-hole pairs should be the main relaxation mechanism for \( s_\downarrow \) and \( p_\uparrow \) states, having lifetimes of 1.3 and 1.0 ns, respectively. For the \( d_\downarrow \) state an additional relaxation is available, namely the \( d_\downarrow \rightarrow p_\uparrow \) transition, which can be radiative \((\Delta m=1)\). Its kinetics determines the first exponent with the 0.8 ns time constant in Fig. 3(a). The expected emission wavelength should amount to 30–50 \( \mu \text{m} \). The second exponent \((1.7 \text{ ns})\) corresponds to the radiative recombination of the \( d_\downarrow \) exciton. The excitation density dependence of the transient PL in Fig. 3(b) reveals the competition of these two relaxation mechanisms. For growing excitation densities the radiative contribution of the \( d_\downarrow \) exciton increases, and its lifetime (second exponent), becomes shorter. In a high-density QDM array this behavior could be explained by a lateral migration of weakly localized \( d_\downarrow \) excitons between neighboring QDMs within the QD plane. Previously, we already observed this effect in dense two-dimensional arrays of QDs with a bimodal size distribution\textsuperscript{10} and in vertically stacked QDs.\textsuperscript{11} In contrast, the contribution of \( p_\uparrow \rightarrow d_\downarrow \) transitions is limited by the \( p_\uparrow \) level occupation. It clearly decreases with the excitation density, and the PL decay time (first exponent) does not show any noticeable changes. At the maximum excitation power \( (2.4 \times 10^{11} \text{ photons}/(\text{pulse} \times \text{cm}^2)) \) the time constants for both exponents become even, and the radiative recombination of \( d_\downarrow \) excitons starts to dominate the scene. Obviously, up to a certain excitation density, the related ratio of \( p_\uparrow \) and \( d_\downarrow \) PL decay times establishes favorable conditions for the \( d_\downarrow \) level population inversion. The restriction to lower densities can be circumvented by a controlled decrease of the QDM density in the array. This should result in a bright emission at wavelengths according to the \( d_\downarrow - p_\uparrow \) split, namely between 30 and 50 \( \mu \text{m} \).

In summary, a consistent interpretation of steady-state PL and transient PL data from quantum dot molecules is presented. The expected exciton lifetime increase is observed. A detailed analysis of the transient luminescence behavior indicates an efficient \( d_\downarrow \rightarrow p_\uparrow \) transition that potentially could be used for the creation of midinfrared light sources.

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