Radiative Recombination Features of Metastable Quantum Dot Array


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Metastable quantum dot (QD) arrays were grown by submonolayer migration enhanced epitaxy (SMEE) mode on GaAs(100) singular and vicinal substrates. It has been established that two QD groups are formed on the vicinal surface. These groups – conjunct QDs (CQD) and isolated QDs (IQD) – demonstrate a basically different behavior in their photoluminescence spectra. The CQDs form chain-like structures. They lay on the wetting layer which connects them to each other. The IQDs are formed due to wetting layer rupture directly on the GaAs surface at the terrace edges. They are isolated and not connected to the rest QD array.

Introduction The morphological stability of QDs which have square base shape and are organized as a dense 2D square lattice array is well established [1, 2]. It has been also shown that for a small area covered by islands the energy gain by forming a square lattice is negligible. As a consequence, a morphologically metastable QD array with another lateral ordering and a different base shape becomes possible [1, 2].

At present, most investigations concerning the QD problem deal with the high-density stable QD array. But it is also known that metastable QDs can show better defined zero-dimensional properties as compared to stable QD arrays [3]. Narrowing of the radiative recombination band down to unprecedentedly low values [4] provides a possibility for device application in spite of their liability to ripening.

Here a question inevitably arises: how can the mechanisms forming QD photoluminescence (PL) be identified? The multimodal structure of QD PL spectra is mainly formed by two mechanisms: recombination through excited states and from QD groups of different origin [5]. Often these mechanisms are relevant at the same time. Thus, their separation from each other is of utmost interest.

Experimental InAs QD layers were grown by SMEE mode. The structures to grow consist of the InAs layer containing QDs confined on both sides by GaAs and Al_{0.25}Ga_{0.75}As/GaAs superlattices (five pairs, 2nm/2nm each). Singular and misoriented

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GaAs(100) substrates towards [001] and [010] directions with 3°, 5°, 7° angles were used. The shutters for In and As were switched on alternatively. In our experiments a 0.5 monolayer (ML) portion of In deposited during each pulse was followed by exposing the surface under As$_4$ flux of 10 s. The growth conditions were maintained the same for all samples. For the InAs QD layer growth the following conditions were set: substrate temperature 470 °C, As$_4$/In effective flux ratio about ten, and growth rate 0.1 ML/s. The surface morphology was controlled in situ by reflected high energy electron diffraction (RHEED). Two kinds of samples were grown: one with mean InAs thickness of 1.8 ML (just after the decomposition of pseudomorphic layer to the QD array) and another with 3 ML, where a well-resolved spotty RHEED pattern is clearly observed. For scanning tunneling microscopy (STM) measurements the samples were immediately quenched after the growth of In-content layers.

Steady-state PL investigations were performed using a Ti-sapphire tuneable laser, pumped by an Ar$^+$ ion laser. The laser beam was focused onto the sample situated in a He cryostat. The luminescence signal was analyzed by a 2 m grating monochromator and detected by a cooled Ge detector.

Transient PL measurements were performed with sub-100 fs excitation pulses (repetition rate of 82 MHz) from Cr:LiSAF and Ti-sapphire lasers at 732 nm (1.69 eV). A grating monochromator in combination with a Streak camera with an infrared-enhanced photocathode allowed spectral discrimination and detection. The total system response is better than 15 ps.

Results and Discussion

STM measurements not shown in this paper revealed the morphological metastability of the resulting QD arrays. The growth mode forms a well-ordered InAs QD array on the singular GaAs surface with chain-like structure along the [001] direction. For 1.8 ML InAs the average QD density is $10^{11}$ cm$^{-2}$ (area coverage 0.3). The QDs have rectangular base shape with lateral sizes of 5 and 12 nm along [011] and [001] directions, respectively. The vicinal surface is characterized by a strong step bunching effect. Smooth terraces are formed between the steps. For a substrate misorientation angle of 7° the average statistical terrace size is 50 nm (theoretically 2.3 nm). QD arrays are located on the terraces. The chain-like structure remains.

PL spectra for different substrate misorientation angles (3°, 5°, 7°) and different InAs layer thicknesses (1.8 ML and 3 ML), measured at 5 K, are shown in Fig. 1. The samples with higher misorientation angle have a QD PL band narrowed and blue shifted. Dramatic changes in the

Fig. 1. PL spectra of InAs QDs grown on GaAs(100) substrates with different misorientation angles in [001] direction; a) 3 ML InAs and b) 1.8 ML InAs. $T = 5$ K, excitation energy $E_{exc} = 1.56$ eV, excitation power density $D = 50$ W/cm$^2$. 
QD PL spectrum take place for high misorientation (7°): the intensive narrow PL band (IS) with a long-wave shoulder arises at a shorter wavelength position, whereas only about 10% of intensity remains at the peak position for the 0° misorientation case (cf. also Fig. 2). In case of 1.8 ML InAs, these changes take place starting from 5° (IS band at 1.37 eV and a residual long-wave shoulder), and in case of 3 ML InAs, starting from 7° (IS band position at 1.34 eV). Such PL spectrum transformation cannot be explained by a simple evolution of the QD array, i.e. decrease of average QD size and deviation from this size.

Figure 2 shows a more resolved PL spectrum of 1.8 ML InAs QDs for 7° misorientation and measured at $T = 77$ K. Deconvolution into Gaussians enables to identify four components within the QD PL spectrum: S0, P0, IS and IP. For S0 and IS bands the time dependence of PL decay is described by one exponent. For P0 this dependence is biexponential (inset in Fig. 2). IP shows a more complex behavior. This is an indicator that only the ground electron state (s-like) and the ground hole state take part in the optical transitions for S0 and IS, whereas P0 and possibly PI describe transitions between ground hole and excited conduction band states. (It should be mentioned here that the wetting layer (WL) emission, whose spectral position is expected at 1.44 eV for $T = 77$ K, practically disappears at $T = 45$ K.)

The dependence of the PL peak energy and the full width at half maximum (FWHM) on power density $D$, spectral composition of the excitation and temperature

![Fig. 2. PL spectrum of 1.8 ML InAs QDs on vicinal surface with 7° [001] misorientation; $T = 77$ K, $E_{exc} = 1.96$ eV. The inset shows the time-resolved PL for this sample. $T = 77$ K, $E_{exc} = 1.69$ eV, $D = 50$ W/cm²]
shows that the pairs S0, P0 and IS, IP belong to two different QD groups with quite different properties, designated as CQD and IQD groups, respectively.

We could prove the absence of carrier exchange between CQD and IQD groups, and the ladder-like mechanism of excitation relaxation within the CQD group by PL excitation spectroscopy [6].

Both an extended Arrhenius analysis made for the QDs in a way similar to that for quantum wells [7] and the independent PL measurement under uniform pressure [8] for the vicinal sample 7° [001] made us to conclude, that P0 and IP are transitions between p-like conduction band state and the ground hole state. The hole activation energy is 50 meV for the CQD group and 40 meV for the IQD.

As already mentioned before, temperature dependences of both PL peak energy and FWHM are basically different for the two QD groups (Fig. 3). In case of the CQD group (S0 and P0) this behavior corresponds to the inter-QD carrier redistribution model [9, 10] extended here for both carrier types. The presence of such a temperature redistribution indicates a close vicinity of CQDs (15 nm from tunneling length calculations [11]). Such QDs are ordered in chain-like structures.

The IS band behavior for increasing temperatures shows the absence of carrier exchange between IQDs. Its FWHM grows and the red shift follows nearly the GaAs band gap change. In this way these IQDs are separated not only from the CQD group but also from each other. As a reason for this we assume the local absence of the wetting layer (WL) due to its rupture at step edges for samples with high degree of step bunching (5°–7°) [12]. Selective excitation in the WL and the GaAs matrix confirms this conclusion [6]. The S0 and P0 bands appear at the resonance excitation of the InAs WL, and the IS band for nonresonant excitation in the GaAs matrix. For these two excitation types the difference between CQD and IQD groups can be clearly observed through the excited QD states.

It has been noted previously [13] that for alternate submonolayer deposition (analogous to SMEE) the chain formation is accompanied by the generation of WL corrugations. So it can be assumed that the interplay of the corrugation structure and the system of steps with high bunching can result in the WL rupture on a vicinal surface. In this way the elastic strain relaxation mechanism, which competes with QD formation, seems to be eliminated, and IQDs are formed.

**Conclusions**  InAs/GaAs metastable quantum dot arrays, grown by SMEE mode on vicinal GaAs substrates, have been studied using STM, steady-state and transient photoluminescence and photoluminescence excitation spectroscopy.
For a high degree of substrate misorientation ($\geq 5^\circ$) it could be concluded that the PL spectrum is formed by two QD groups: the connected QDs are included in chain-like structures. They lay on the wetting layer (WL) and are connected with each other by means of it. As a result carrier exchange becomes possible. On the contrary, isolated QDs are formed at terrace edges due to the WL rupture. They are not connected to the QD array, and carrier exchange is not possible for them.

A more comprehensive analysis of the contribution of both conjunct and isolated QDs to spectra formation will be given elsewhere [6].

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