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Structure and optical anisotropy of vertically correlated submonolayer InAs/GaAs quantum dots

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A vertically correlated submonolayer (VCSML) InAs/GaAs quantum-dot (QD) heterostructure was studied using transmission electron microscopy, high-resolution x-ray diffraction (HRXRD) and polarization-dependent photoluminescence. The HRXRD (004) rocking curve was simulated using the Tagaki-Taupin equations. Excellent agreement between the experimental curve and the simulation is achieved assuming that indium-rich VCSML QDs are embedded in a quantum well (QW) with lower indium content and an observed QD coverage of 10%. In the VCSML QDs, the vertical lattice mismatch of the InAs monolayer with respect to GaAs is around 1.4%, while the lattice mismatch in the QW is negligible. The photoluminescence is transverse magnetic—polarized in the edge geometry. © 2003 American Institute of Physics. [DOI: 10.1063/1.1581005]

Much interest has been attracted to the growth of self-assembled semiconductor quantum dots (QDs), due to their potential application in optoelectronic devices.¹ Submonolayer (SML) deposition is an alternative method to the widely used Stranski–Krastanow (SK) mode of growing QDs.^{2–4} Deposition of a SML InAs on a GaAs (001) surface leads to the formation of 1 ML high InAs islands elongated along the $[1\bar{1}0]$ direction.² The density and the lateral sizes of the islands depend on the deposition amount. Vertical correlation of the SML QDs occurs when stacking the SLM QDs with thin spacers.^{5–9} High-power lasers with the stacked SML InAs/GaAs QDs as the active region have recently been demonstrated.^{6–8} The structure and optical properties of the SK InAs/GaAs QDs have been intensively studied,¹ but few papers have been reporting on the vertically correlated (VC) SML InAs/GaAs QDs.^{4,9} In this letter, the structure and the optical anisotropy of the VCSML InGaAs QDs are investigated by plan-view transmission electron microscopy (TEM), high-resolution x-ray diffraction (HRXRD), and polarization-dependent photoluminescence (PL) at low temperature in both the backscattering and the edge emission geometries.

The VCSML InAs/GaAs QD sample was molecular-beam epitaxy grown on a semi-insulating GaAs (001) substrate. After oxide desorption, a 500 nm GaAs buffer layer, 20 nm AlAs, and 82 nm GaAs were grown at 600 °C. After the substrate temperature was lowered to 480 °C, 10 cycles of InAs(0.5ML)/GaAs(2.5ML) were deposited to form a VC-SML QD layer, with 2 nm of GaAs covering the VCSML QD layer. Then, the substrate temperature was again in-

creased to 600 °C to grow 20 nm of AlAs and a 106 nm GaAs cap layer.

TEM studies were performed using a Philips CM20 instrument operating at 200 keV. Figure 1 shows a plan-view TEM image of the VCSML InAs/GaAs QDs. The contrast is mainly due to strain fields. 10% of the surface is covered by QDs and the area density of QDs is around $5.2 \times 10^{11} \text{ cm}^{-2}$, much higher than the conventional SK InAs/GaAs QDs. Most of the QDs are slightly elongated along $[1\bar{1}0]$ direction, and the size of QDs is around 5–10 nm in diameter. However, the actual QD size could be smaller since the strain field may extend beyond the QD boundary.

The determination of the strain in the QDs is useful not only for understanding the QD growth but also for the calculation of the energy diagram of the QDs.^{5,10,11} HRXRD is

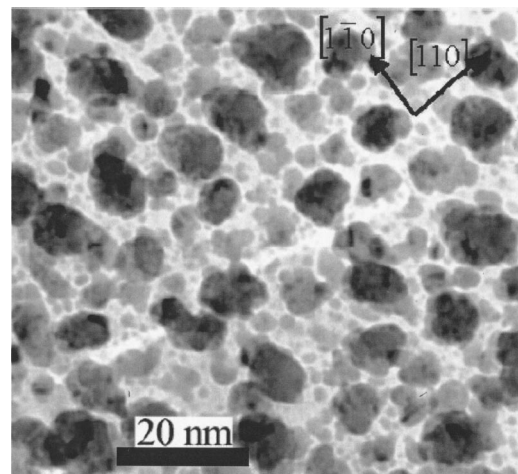


FIG. 1. (011) bright-field plan-view TEM image of the structure with VCSML InAs/GaAs QDs.

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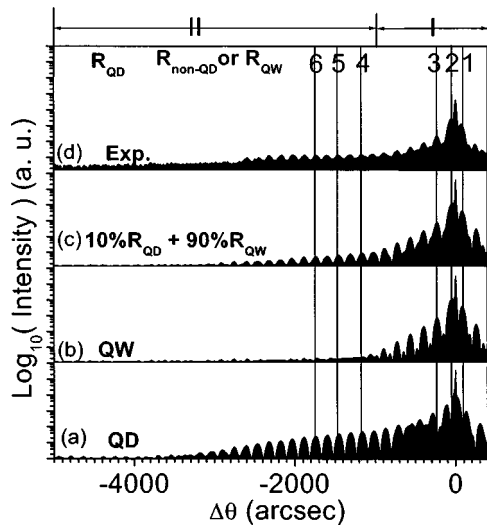


FIG. 2. HRXRD rocking curves in the vicinity of the GaAs (004) reflection, (a) the simulated curve of R_{QD} , assuming $a_{\text{InAs}}^{\perp} = 5.7324 \text{ \AA}$, (b) the simulated curve of R_{QW} , assuming $a_{\text{In}_{0.15}\text{Ga}_{0.85}\text{As}}^{\perp} = 5.6533 \text{ \AA}$, and (c) the combination of (a) and (b), assuming the QD coverage percentage is 10%, and (d) the experimental data. The curves are shifted vertically for clarity. The inset shows the different contributions from the QD parts and the lateral non-QD parts to the reflection coefficient of a VCSML QD sample. Lines 1 to 6 are guides for the eyes.

an important nondestructive method of determining the interface morphology and the strain of the buried films. The measured rocking curves are the “fingerprints” of the investigated structure and can be analyzed by using the dynamical x-ray diffraction theory, i.e., the Tagaki–Taupin equations.^{11,12} Due to the structural complexity of a QD heterostructure, the average strain of the QD plane rather than of the QDs themselves has been determined for the SK QDs.^{13–15} In the case of the VCSML QDs, the contribution from the QD parts and the lateral non-QD parts to the finally measured reflection rocking curve can be considered separately, as shown in the inset of Fig. 2. According to the growth mechanism of stacked SML InGaAs QD heterostructure,⁷ the VCSML QD in our sample can be described as 10 cycles of GaAs(2ML)/InAs(1ML), while the lateral non-QD part can be described as a 30 ML GaAs and 50% of the surface should be covered by the VCSML QDs. However, QDs only fill about 10% of the surface in the plan-view TEM image (Fig. 1). This means that 40% of the SML InAs is in the lateral non-QD part, possibly due to In–Ga intermixing or because vertical correlation does not occur for all the SML QDs. Therefore, the lateral non-QD part is an InGaAs alloy rather than pure GaAs, and our sample is a QD–quantum-well (QW) heterostructure. The average In composition in the lateral QW is calculated to be around 15%, according to the 10% QD coverage on the surface.

The HRXRD rocking curve near the (004) reflection peak of GaAs (001) substrate was measured using a MAC Science HRXRD instrument with a Cu $K\alpha$ x-ray source and a Ge (004) monochromator. Figure 2 shows the experimental data and simulated results based on Tagaki–Taupin equations. The total reflection coefficient $R(\Delta\theta)$ of the sample can be written as

$$R(\Delta\theta) \approx xR_{\text{QD}}(\Delta\theta) + (1-x)R_{\text{QW}}(\Delta\theta), \quad (1)$$

where $\Delta\theta$ is the angular deviation from the Bragg angle of the (004) peak of the GaAs substrate, x is the QD coverage percentage on the surface, $R_{\text{QD}}(\Delta\theta)$ and $R_{\text{QW}}(\Delta\theta)$ are the reflection coefficients from the part of the sample with only the VCSML QDs and the part with only the lateral QW, respectively. Note that Eq. (1) is valid only when $xR_{\text{QD}}(\Delta\theta)$ is so small that the multiple scattering of x rays between the QD and the QW parts is negligible. In the angular range from -1000 to $+500$ arcs (range I), the reflectivity is much higher than outside this range and mainly depends on the $R_{\text{QW}}(\Delta\theta)$ because QW covers 90% of the surface. Only when the lattice mismatch is negligible between the lateral 8.48 nm $\text{In}_{0.15}\text{Ga}_{0.85}\text{As}$ QW and GaAs, do the calculated interference fringes [curve b in Fig. 2(b), lines 1 to 3] match the experimental ones in range I. At the same time, no interference fringes in the range from -1000 to -4000 (range II) in curve b can be comparable to the experimental data. Therefore, the interference fringes in range II are mainly from $R_{\text{QD}}(\Delta\theta)$. By varying the lattice constant a_{InAs}^{\perp} of the InAs monolayer inside the VCSML QDs, $R_{\text{QD}}(\Delta\theta)$ is calculated and found to match the interference fringes [curve a in Fig. 2(a), lines 4 to 6] only when $a_{\text{InAs}}^{\perp} \approx 5.7324 \text{ \AA}$, i.e., a_{InAs}^{\perp} is around 1.4% higher than the lattice constant of GaAs. With $x = 10\%$ determined from the TEM image (Fig. 1), curve c in Fig. 2(c) is calculated from Eq. (1). It can be seen that all the interference fringes in curve c in Fig. 2(c) can match the experimental ones very well. The macroscopic continuum elasticity theory (MCET) predicts that for a pseudomorphic InAs layer buried in GaAs (001), the strain normal to the (001) plane would be 7.26%, corresponding to $a_{\text{InAs}}^{\perp} \approx 6.4981 \text{ \AA}$.^{16,17} However, the validity of the MCET in the monolayer limit is a long debated issue.^{18–20} In the case of SML InAs QDs, the InAs islands with 1 monolayer in height and around 5–10 nm in diameter are surrounded by a GaAs matrix, and the MCET cannot be applied directly. Our HRXRD gives an experimental determination of the strain in the SML InAs QDs.

Shape anisotropy effects on the electronic properties of VCSML InAs/GaAs QDs are investigated here by measuring the polarization dependence of the optical transitions. The PL was excited at 10 K by a He–Ne laser at 632.8 nm with an excitation density $\sim 10 \text{ W/cm}^2$. A 64 cm monochromator with a Si charge coupled device was used to detect the PL. The polarization of the PL was analyzed using a fixed polarizer and a broadband $\lambda/2$ plate. The PL polarization anisotropy is defined as $P = (I_{\parallel} - I_{\perp}) / (I_{\parallel} + I_{\perp})$, where I_{\perp} is the vertically polarized intensity and I_{\parallel} is the horizontally polarized intensity in the laboratory coordinate system.

Figure 3 shows the degree of linear polarization of PL detected in both the backscattering [curve (a) in Fig. 3(a)] and the edge geometries [curves (b) and (c) in Figs. 3(b) and 3(c), respectively]. In the backscattering geometry, the light propagates in the [001] direction (surface emission), I_{\parallel} and I_{\perp} are polarized along the $[1\bar{1}0]$ and $[110]$ directions, respectively. The broad peak [curve (d) in Fig. 3(d)] centered at 1.26 eV with a linewidth of 55 meV is assigned to the ground-state transitions of the VCSML InAs/GaAs QDs. PL emission from the QDs is predominantly polarized along the $[1\bar{1}0]$ direction in the whole energy range, $P \approx 0.22$ [curve (a) in Fig. 3(a)], due to the elongation of the QDs along the

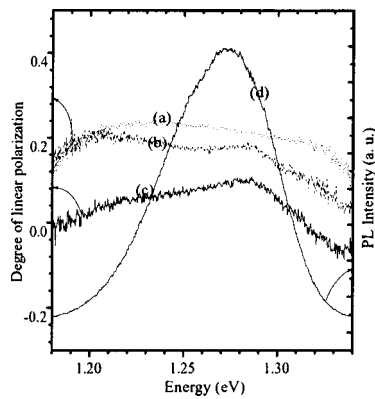


FIG. 3. The degree of linear polarization of PL measured at 10 K, (a) in the backscattering geometry, collecting light in the $[001]$ direction, (b) in the edge geometry, collecting light in the $[1\bar{1}0]$ direction, and (c) in the edge geometry, collecting light in the $[110]$ direction. Curve (d) is a PL spectrum measured in the backscattering geometry, polarized along the $[1\bar{1}0]$ direction.

$[1\bar{1}0]$ direction, as shown in the plan-view TEM observation.

In the edge geometry, the spectra were obtained exciting and detecting at the sample edges, the light propagates in $[110]$ or $[1\bar{1}0]$ direction, I_{\parallel} and I_{\perp} are polarized along the $[001]$ and $[1\bar{1}0]$ or $[110]$ directions, respectively. These polarizations are, respectively, referred to as transverse magnetic (TM) and transverse electric (TE) modes. To suppress the PL emission from the $[001]$ sample surface, the edge emission was spatially selected in near and far field. From curves (b) and (c) of Figs. 3(b) and 3(c), respectively, the PL emission was found to be TM polarized in the VCSML InGaAs/GaAs QDs, while a single-layer InGaAs/GaAs SK QD structure shows a TE polarization, measured in the same geometry and using similar equipment. This indicates that the electronic states are elongated along the growth direction for VCSML InGaAs/GaAs QDs. A similar result has been observed in the case of vertically coupled CdSe/ZnS SML QDs and InGaAs/GaAs SK QDs.^{4,21} A slightly higher anisotropy of $P \approx 0.17$ for emission along the $[1\bar{1}0]$ direction [curve (b) in Fig. 3(b)] than that of $P \approx 0.09$ along the $[110]$ direction [curve (c) in Fig. 3(c)] is due to the fact that the lateral dimensions of VCSML QDs along $[1\bar{1}0]$ direction are nearer to the QD heights. This feature is not only of the fundamental interest for electronic structure and optical properties, but also important in its applications in polarization-independent devices.

In summary, the structure and optical anisotropy of VCSML InAs/GaAs QDs, are studied using plan-view TEM, HRXRD, and polarization-dependent PL at low temperature in both the backscattering and edge geometries. The method of calculating the reflection coefficient from a VCSML QD

heterostructure was demonstrated, using the Tagaki-Taupin equations, the structure of VCSML QDs, and the QD coverage percentage on the surface. The separate contributions from the QDs and the lateral QW to the total x-ray diffraction signal have been extracted. The lattice mismatch in the InAs monolayer inside the VCSML is found to be around 1.4% with respect to GaAs, and that in the lateral QW is negligible. In the edge geometry, the PL emission is TM polarized, which is important for the application of VCSML QDs in optoelectronic devices.

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