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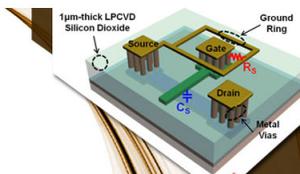Information for Authors: http://apl.aip.org/authors

Highly polarized self-assembled chains of single layer InP/(In,Ga)P quantum dots

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Well-ordered chains of InP quantum dots on a self-organized undulating $In_{0.48}Ga_{0.52}P$ surface exhibit optical anisotropy due to their shape and the $In_{0.48}Ga_{0.52}P$ matrix. The structures are investigated by polarization-dependent photoluminescence together with transmission electron microscopy. Luminescence from the $In_{0.48}Ga_{0.52}P$ matrix is polarized in one crystallographic direction due to anisotropic strain arising from a lateral compositional modulation. The photoluminescence measurements show enhanced linear polarization in the alignment direction of quantum dots, $[\bar{1}10]$. A polarization degree of 66% is observed; this polarization in emission is independent of the polarization of the excitation laser. The optical anisotropy is achieved with a straightforward heterostructure, requiring only a single layer of QDs. © *2010 American Institute of Physics.* [doi:10.1063/1.3529467]

Semiconductor quantum dots (QDs) grown via the Stranski–Krastanow mechanism are interesting both for fundamental physics as well as for device applications, due especially to the simplicity of their growth and to their good crystal quality. Controlling the position of QDs is particularly important for device applications, for instance, in lasers[1,2] or in computing.[3–5] Control of polarization direction is also important for vertical-cavity surface-emitting lasers (VCSELs) to control the polarization mode without an external polarizer.[6] Sophisticated techniques have been recently developed to obtain self-assembled highly ordered QD structures that need stacking layer of QDs and/or surface miscut angles; see, e.g., Refs. 7 and 8. A more recent example of ordered self-organized growth of QDs describes laterally ordered $InP/In_{0.48}Ga_{0.52}P$ QDs on GaAs along the $[\bar{1}10]$ crystallographic direction by using a self-organized undulating $In_{0.48}Ga_{0.52}P$ buffer layer, lattice-matched to a GaAs substrate, as a template.[9] The impact of the InGaP layer is analyzed with the help of grazing-incidence x-ray diffraction and photoluminescence measurements in a previous paper.[9]

In this letter, the polarization behavior of an undulated InGaP buffer layer and the InP QDs that are formed on top of the undulations is investigated. The undulations of the InGaP layer, grown on a GaAs substrate with a miscut smaller than 0.05°, are used as a template for the aligned QD growth in the $[\bar{1}10]$ direction. The samples are also investigated by transmission electron microscopy. These measurements address the mechanism for the formation of the undulations, whether lateral composition modulation (LCM) or long range ordering (LRO) takes place in the InGaP layer and is responsible for the formation of the InGaP undulations. At the same time the effects of these mechanisms (LCM and LRO) on QDs that are formed on the ridge of the undulations are investigated through polarization-dependent photolumi-

nescence (PL) measurements that give information about electronic energy levels. Polarization of the emitted light contains additional information about the electronic states, both about the matrix within which the QDs are embedded as well as about the lifting of degeneracies in QDs due to mechanisms such as LCM or LRO. Both LCM and LRO should cause a smaller band gap. Because of valence band splittings, a partial polarization in the $[\bar{1}10]$ direction is predicted for LCM. Polarization in the $[110]$ direction, however, is expected in the case of LRO.[10,11] Thus, polarization-dependent photoluminescence may indicate which mechanism takes place.

The self-assembled laterally ordered chains of InP QDs are grown using gas-source molecular beam epitaxy (GSMBE) on an $In_{0.48}Ga_{0.52}P$ buffer layer on a GaAs (001) substrate. The growth conditions and mechanisms for obtaining laterally ordered chains of QDs are discussed in Ref. 9. In that work, we observed that growing the InGaP buffer layer, lattice-matched to GaAs, at higher substrate growth temperatures (470 °C) causes undulations along $[\bar{1}10]$ that act as templates for the growth of laterally aligned InP QDs. The alignment of the QDs is caused by an ordered strain field, which results from the preferential surface diffusion of In adatoms during the growth of the $In_{0.48}Ga_{0.52}P$ buffer layer. In this paper, we first focus on the optical properties of the InGaP buffer layer where the QDs are embedded and then on the effect of this matrix on the optical properties of laterally ordered QDs. An enhanced polarization of PL is observed in the $[\bar{1}10]$ compared to the $[110]$ direction, pointing toward LCM as the responsible mechanism. The possible mechanisms for the observed enhanced polarization are described.

The structures were grown in a RIBER-21T GSMBE system. After oxide desorption, a 100 nm thick GaAs buffer was grown at 550 °C at a rate of 0.39 ML/s, followed by 230 nm of $In_{0.48}Ga_{0.52}P$ grown at 470 °C. Subsequently, 0.4
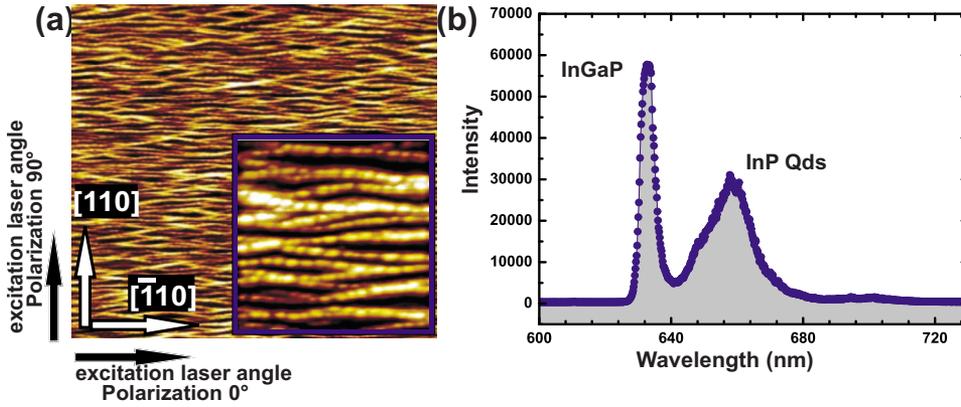
FIG. 1. (Color online) (a) AFM micrograph of a sample without a cap layer with excitation directions, the height contrast is between 0 and 6 nm, the scale for AFM micrograph is $3 \times \mu m^2$, and the scale for inset micrograph is $0.5 \times \mu m^2$. (b) PL spectrum for laterally ordered QDs; emission at 630 nm stems from InGaP and emission at 660 nm stems from InP QDs.

ML of InP for the QDs was deposited at 410 °C using an ultralow growth rate of 0.015 ML/s. Finally, a capping layer of In$_{0.48}$Ga$_{0.52}$P with thickness of 100 nm is grown (sample A). Using such a low deposition rate for the InP may encourage the formation of the QDs only on the hills where the lattice can be more easily deformed compared to the valleys [Fig. 1(a)]. At lower growth rates the migration length of In adatoms increases. Larger migration lengths allow the formation of QDs whose position and size are driven by the strain and surface energies.[12,13]

Control structures without undulations were also prepared using the same growth conditions, except that the In$_{0.48}$Ga$_{0.52}$P growth temperature was lower (440 °C). Sample B discussed in this work is grown with such an In$_{0.48}$Ga$_{0.52}$P buffer without undulations and without InP QDs. Reflection high-energy electron-diffraction patterns were monitored *in situ* during growth, and the surface of In$_{0.48}$Ga$_{0.52}$P shows a (2×1)-reconstruction for all samples. Investigating several InGaP samples grown at different substrate growth temperatures (in the range 440–500 °C) by atomic-force microscopy shows that undulations that will serve as templates for laterally aligned QDs form only in a specific window of growth conditions with growth temperature near 470 °C. Increasing the temperature to 490 °C results in the InGaP surface growing islandlike, while decreasing the temperature to 440 °C results in a smooth surface, upon which homogeneously distributed QDs can be grown.[14]

Polarization-dependent microphotoluminescence (μ-PL) measurements were performed using an objective with numerical aperture of 0.7 that focuses the above-bandgap exci-

tation illumination from a 532 nm laser onto the sample and also collects the emitted luminescence. A μ-PL spectrum of the laterally ordered InP/InGaP QD sample is shown in Fig. 1(b). The shorter wavelength luminescence peak centered at 630 nm originates from the InGaP buffer layer and the longer wavelength luminescence peak, centered at 660 nm, from the InP QDs. To examine the polarization properties of each spectral feature, Fig. 2 depicts the μ-PL intensity as a function of collection polarization angle (θ_c) for three different excitation laser polarizations (θ_e) from two different samples. Sample A has InGaP undulations and sample B does not. As illustrated in Fig. 1(a), the $[\bar{1}10]$ direction defines the 0° linear polarization direction for both excitation and collection polarization. In Fig. 2(a) the dependence of the collection polarization from the InGaP buffer layer of sample A [630 nm peak in Fig. 1(b)] is depicted for $\theta_e=0°$, 45°, and 90°. Figure 2(b) shows results from the same measurement for the InP QDs, also of sample A [660 nm peak in Fig. 1(b)]. In both Figs. 2(a) and 2(b), the InP QDs and InGaP buffer layer collection polarization are independent of the excitation laser polarization. From the data in Fig. 2 it is possible to determine the degree of linear polarization, which is defined as

$$P = \frac{I_{\theta_{c=0}} - I_{\theta_{c=90}}}{I_{\theta_{c=0}} + I_{\theta_{c=90}}}, \quad (1)$$

where $I_{\theta_{c=0}}$ is the (μ-PL) intensity along θ_c. The degrees of polarization are 46% for the InGaP buffer layer and 66% for the InP QDs, both from sample A. Figure 2(c) depicts the
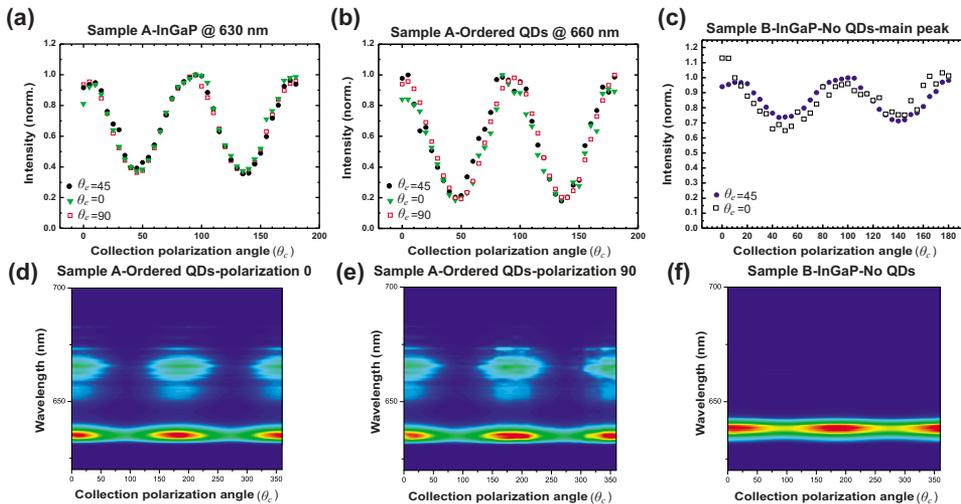

FIG. 2. (Color online) Polarization-dependent PL measurements for the samples with QDs. (a) The collection polarization dependence of the InGaP buffer layer, in sample A, for $\theta_e=0°$, 45°, and 90°. (b) The collection polarization dependence of the InP QDs, in sample A, for $\theta_e=0°$, 45°, and 90°. (c) The collection polarization dependence of the InGaP buffer layer, in sample B, for $\theta_e=0°$ and 45°; $\theta_e=0°$ is normalized at 10° collection polarization since the data points for $\theta_e=0°$ and 5° were noisy. (d) Two-dimensional (2D)-plot for excitation polarization direction $\theta_e=0$, sample A. (e) 2D-plot for excitation polarization direction $\theta_e=90$, sample A. (f) 2D-plot for excitation polarization direction $\theta_e=0$, sample B.

collection polarization dependence of the InGaP buffer layer for sample B; the degree of linear polarization for the InGaP buffer layer is 16%, in contrast with the data of sample A depicted in Fig. 2(a) and reflecting the influence the undulations have on the (μ-PL) polarization orientation.

The difference in the degree of polarization of the PL emitted from the InGaP buffer layer for samples A and B (46% and 16%, respectively) may be explained by two different effects. One of them is atomic ordering. If there is a CuPt-type ordering in the system, a double-periodic crystal-field is introduced and the lattice periodicity in the $[\bar{1}11]$ doubles (from zincblende structure with cubic symmetry to a CuPt–B structure with trigonal symmetry). The resulting splitting of the otherwise degenerate states may enhance the degree of polarization. This kind of ordering favors the transition of light polarized along the [110] direction. On the other hand, if there is lateral composition modulation in our system, the degeneracy at the top of valence band is lifted, favoring optical transitions polarized along the $[\bar{1}10]$ direction.[15] If one of these properties is weak in the structure then the anisotropy is more pronounced in the opposite direction. The enhanced intensity in the $[\bar{1}10]$ direction leads us to conclude that LCM is dominant in our structure and may be the reason for the formation of undulations.

The PL spectra were independent of the excitation laser polarization, indicating that the excited electron-hole pairs do not retain any polarization by the time they recombine. The polarization of the 660 nm emission from the InP QDs may result from the anisotropic shape of InP QDs, as suggested earlier.[16] On the other hand, the anisotropic matrix, within which the QDs are embedded, causes anisotropic strain. We have previously shown that the observed undulations have a larger horizontal lattice spacing in the [110] direction.[9] The horizontal and vertical strains in the undulation regions relative to the GaAs substrate have been evaluated as $\varepsilon_{\parallel} \equiv \varepsilon_{110} = 1.26 \times 10^{-3}$ and $\varepsilon_{\perp} = 2.5 \times 10^{-4}$ inside the buffer layer, respectively.[9] This strain anisotropy can also directly cause an anisotropy in the polarization of the optical emission, independent of the structural morphology.[17] We know from previous works that the aligned QDs have polarization degree of 36%–45% with optical anisotropy for both ordered and homogeneously distributed QDs.[15–17] The degree of polarization for the QDs on the undulations (66%) is, however, higher than the 36%–45% for QDs without undulations. This enhanced polarization could be due to the additional strain resulting from the undulations, or could be due to the valence band splittings associated with LCM or LRO. Radiative recombination in these states may exhibit differing polarizations according to the appropriate selection rules. Fine splitting of the degenerate quantum dot states into components containing [110]-allowed, $[\bar{1}10]$-allowed, and [001]-allowed optical transitions is expected.[10]

High resolution transmission electron microscopy (TEM) measurements were also performed on the samples with and without undulations. As shown in Fig. 3, in the reference sample grown at 440 °C (without undulations), there is no compositional modulation since the image appears to be smooth. However, in the sample grown at 470 °C (with undulations), composition modulation is observed, with In-rich (darker regions) and Ga-rich (brighter regions)
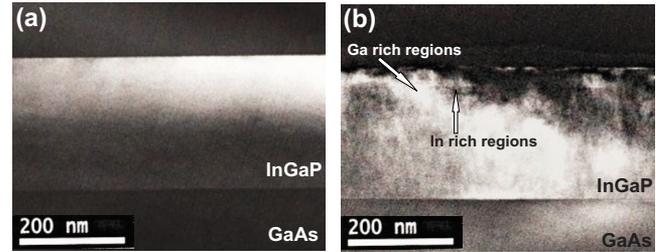

FIG. 3. (Color online) TEM images for the samples (a) without undulations, no lateral composition can be seen; and (b) with undulations, lateral composition can be seen with darker regions to be In-rich and brighter regions to be Ga-rich.

regions. New TEM measurements are planned to obtain direct information about atomic ordering as well as extended x-ray absorption fine structure measurements to directly investigate the related atomic bonds.

In conclusion, we have demonstrated that embedding the laterally ordered QDs into a matrix, which has an undulating surface, changes the intrinsic properties of QDs. This new environment for QDs enhances the linear polarization of the optical emission. For device applications such as for VC-SELS, it is important to control the polarization and its direction without the need of a polarizer. We expect that the ability to achieve optical anisotropy with a straightforward heterostructure, requiring only a single layer of QDs without stacking layers or surface misorientation, will lead to further investigations and also to applications.

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