Influence of quantum-dots density on average in-plane strain of optoelectronic devices investigated by high-resolution X-ray diffraction

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High-resolution X-ray diffractometry is used to probe the nature of a diffraction-peak broadening previously noticed in quantum dots (QDs) systems with freestanding InAs islands on top of GaAs (001) substrates [Freitas et al., Phys. Stat. Sol. (a) **204**, 2548 (2007)]. The procedure is hence extended to further investigate the capping process of InAs/GaAs QDs. A direct correlation is established between QDs growth rates and misorientation of lattice-planes at the samples surfaces. This effect provides an alternative tool for studying average strain fields on QDs systems in standard triple axis diffractometers running on X-ray tube sources, which are much more common than synchrotron facilities.

1 Introduction Self-organized quantum dots (QDs) on semiconductor surfaces are typically used as light-emission sources in many optoelectronic devices [1]. Due to processing requirements, these nanostructures have to be covered by a few layers of another material; in general the same material of the substrate. QDs density, size, shape, and composition are fundamental parameters in determining their optoelectronic properties. These parameters are influenced, before capping, by the QDs growth rates. Most efforts carried out in the last few years have been focused on understanding what happens to the QDs after capping since it provides the final tailoring in the device's properties [2–12]. However, how these structural parameters affect the capping process is a new subject, whose systematic investigation requires a technique capable of accessing average structural information on large ensembles of embedded QDs. Recently [13], X-ray Renninger scanning has shown that under certain QDs growth conditions, high levels of in-plane stresses appear in the InAs/GaAs (001) QD system after capping. Moreover, it has also shown that the rocking-curve intensity profile of the GaAs 002 reflection is sensitive to the presence of freestanding QDs as well as to the capping process itself. In this work, high-resolution X-ray diffractometry is employed in an attempt to understand how a weak, low-attenuating reflection, such as the GaAs 002, can be sensitive to what is going on into a few tens of nanometers at the sample surface. The observed effect is hence exploited to investigate the capping process in a series of samples differing only by the QDs growth rates.

2 Experimental Samples were MBE grown on semi-insulating GaAs (100) substrates, with 200 nm GaAs buffer layers deposited at 580 °C [14]. QDs were formed by deposition of 2.4 monolayers (ML) of InAs at 510 °C and using deposition rates τ, ranging from 0.0055 ML/s to 0.10 ML/s, as summarized in Table 1. A 30 nm GaAs cap layer was grown at the same substrate temperature of the QDs, 510 °C. High-resolution X-ray diffraction measurements were performed on a Philips X'Pert-MRD high resolution diffractometer: Cu tube, 4-crystal asymmetric 220 Ge monochromator and 3-bounce 220 Ge analyzer crystal. Nominal spectral width $\Delta\lambda/\lambda = 2 \times 10^{-4}$. X-ray beam divergences are

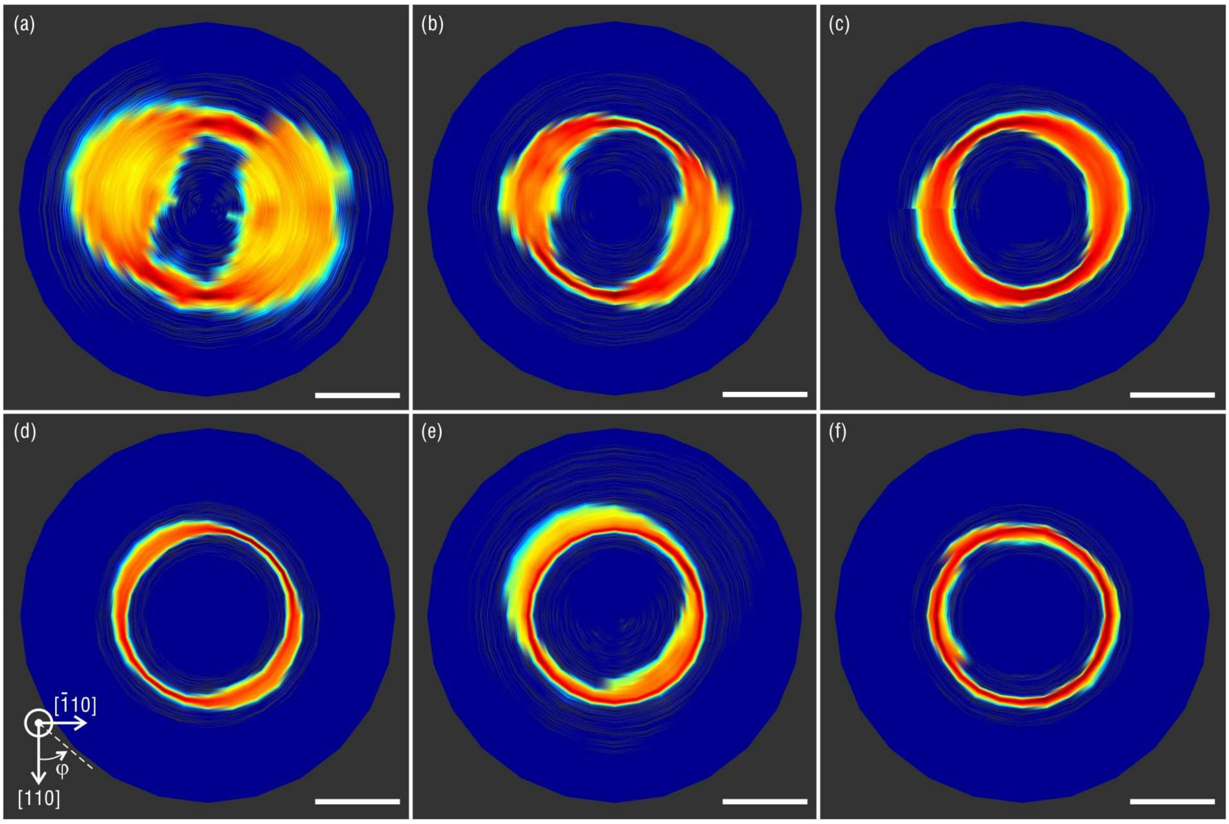

Figure 1 Pole diagrams of the GaAs 002 reflection in triple-axis goniometry for a series of samples: (a) with exposed QDs (no cap-layer) and $\tau = 0.09$ ML/s; (b-f) with embedded QDs, 30 nm thick cap-layer, and (b) $\tau = 0.0055$ ML/s, (c) $\tau = 0.0092$ ML/s, (d) $\tau = 0.031$ ML/s, (e) $\tau = 0.049$ ML/s, and (f) $\tau = 0.10$ ML/s. Each diagram contains 24 transversal scans of the reciprocal space as a function of the azimuthal φ angle, while each transversal scan corresponds to the diffracted intensity across the center of the 002 reciprocal lattice point where $Q_z = 2/a_{\rm GaAs} = 3.5377\,{\rm nm}^{-1}$, i.e. at $2\theta = 31.6264°$ from the incident beam direction; analyzer acceptance angle is $12''$. Scale bars stand for $5 \times 10^{-3}\,{\rm nm}^{-1}$ in reciprocal space units (r.s.u.) or $291.5''$ in angular units of the sample's rocking-curve angle.

$12''$ and $2°$ in the diffraction plane and in the axial direction, respectively. During data acquisition in this system, the diffraction vector of a chosen single-crystal reflection is repositioned back onto the diffraction plane of the diffractometer every time the sample undertake an azimuthal rotation.

3 Results and discussions In a previous investigation, rocking-curves of the GaAs 002 reflection carried out on sample #6 as a function of its azimuthal φ rotation has shown an unexpected broadening at certain azimuths [13]. Since the measurements had been performed on a synchrotron station with an open detector, either lattice strain or misorientation of the diffracting planes could be responsible for such broadening. A similar measurement on this sample is shown in Fig. 1(a) with the difference that, at this time, the diffracted beam direction has been selected by the narrow acceptance, $12''$, of the analyzer crystal. Hence, only unstrained GaAs lattice planes, regarding the instrumental resolution of $\Delta a/a \simeq 4 \times 10^{-4}$, are contributing

Table 1 Samples with QDs formed by deposition of 2.4 ML of InAs using different deposition rates, τ. QDs density prior to capping estimated by linear interpolation of values (*) obtained from atomic force microscopy in samples #2 and #6 with exposed QDs [13]. The average volume of InAs material per QD, $V_{\rm QD}$, is calculated by considering 0.325 nm as the monolayer thickness of strained InAs.

Sample (#)	τ (ML/s)	Density (QDs/μm^2)	$V_{\rm QD}$ ($\times 10^3$nm^3)	Cap-layer (30 nm)
1	0.0055	174	4.5	$\times 1$
2	0.0070	177*	4.4	$\times 0$
3	0.0092	182	4.3	$\times 1$
4	0.031	230	3.4	$\times 1$
5	0.049	270	2.9	$\times 1$
6	0.09	360*	2.2	$\times 0$
7	0.10	382	2.0	$\times 1$

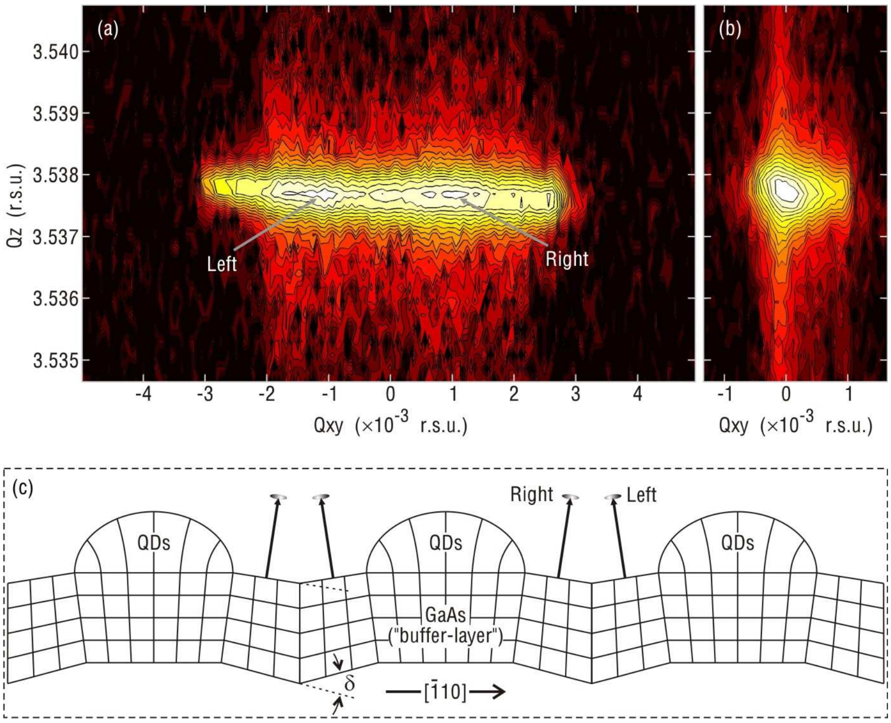

Figure 2 (a,b) Reciprocal space maps of the GaAs 002 reflection carried out on sample with exposed QDs ($\tau = 0.09\,$ML/s) and at two perpendicular azimuths: (a) $\varphi = 84.3°$ and (b) $\varphi = -5.7°$ regarding the [110] crystallographic axis. (c) Freestanding QDs as responsible for an elastic-strain modulation along a single [110] type of in-plane direction in the GaAs buffer-layer lattice, as suggested by the observed preferential misorientation of the 002 reciprocal vector, see also the pole diagram in Fig. 1(a). By taking $\Delta Q_{\rm xy} \simeq 2.18(\pm 0.05) \times 10^{-3}\,{\rm nm}^{-1}$ divided by $Q_{\rm z} = 3.5377\,{\rm nm}^{-1}$, $\delta = 0.035°$ corresponds to the misorientation angle between the left and right intensity maxima in the map (a). Experimental setup: CuKα_1 in triple-axis goniometry. r.s.u. = nm^{-1}.

to the intensity data. It implies that a preferential misorientation of the (002) diffracting planes is the most probable cause of the observed broadening. In Figs. 1(b-f), the same type of measurement, called pole diagram, on samples with capped QDs also show this preferential misorientation effect, although in a smaller magnitude that decreases even more for increasing QDs deposition rates.

From the extinction depth definition of the dynamical theory of X-ray diffraction (textbooks, e.g. Ref. [15]), 50% attenuation of the incident beam intensity occurs in 4.1 μm for the GaAs 002 reflection when using CuKα_1 radiation. Highly-strained lattice regions, above the instrumental resolution (of 4×10^{-4}), are expected to exist locally at the QDs interfaces within the surrounding GaAs matrix (buffer and cap layers), and into a total depth not larger than the cap layer thickness, i.e. a few tens of nanometers [11]. Reciprocal space maps of the 002 reflection at several azimuths, e.g. Figs. 2(a,b), have evidenced that lattice strain is not stretching the 002 reciprocal lattice point along the surface normal direction, and hence it is not causing the observed changes in the line-profile intensities of this reflection. On the other hand, an angular misorientation of $\delta = 0.035°$ in the diffraction vector, generating the left and right intensity maxima in Fig. 2(a), can be achieved by a very tiny strain modulation below 10^{-4}, or even less, as schematically depicted in Fig. 2(c).

A much shorter probing depth for this strain modulation is obtained by using the 004 reflection. It is 10 times shorter than in the case of the 002 reflection, i.e. 50% intensity attenuation occurring in 0.4 μm. As seen in Fig. 3, the reciprocal space map of the 004 reflection, at the same azimuth of the 002 map in Fig. 2(a), also shows two intensity maxima split by $\Delta Q_{\rm xy} \simeq 2.55(\pm 0.15) \times 10^{-3}\,{\rm nm}^{-1}$, nearly the same value of $2.18(\pm 0.05) \times 10^{-3}\,{\rm nm}^{-1}$ obtained in the 002 map. It implies in a lattice misorientation of $\delta = 0.021°$. Measurement of a smaller δ value when probing closer to the surface is in agreement with our hypothesis of strain modulation, Fig. 2(c), since it is caused by the constrain of the substrate lattice that prevents the sample to be bent freely in response to the in-plane strain introduced at the bottom of the QDs. Another hypothesis would be a shortening of the lateral (in-plane) lattice coherence as responsible for the transversal broadening, by nearly a same amount, of both reciprocal lattice points. However, this later hypothesis has been ruled out by the occurrence of the two distinct intensity maxima instead of a single broad maximum on each reciprocal space map.

Although a symmetric Bragg reflection is unable to directly detect the strain fluctuation at the QDs, it seems indeed capable of probing lattice misorientation induced by a weak strain field that is generated at the QDs lattice mismatched interfaces and propagated several microns below the surface into the GaAs substrate lattice. The misorientation occurs preferentially towards one in-plane direction only, probably because the low strain energy is not enough to produce another similar pattern of strain modulation along a perpendicular direction of the one that is already established. In samples with exposed QDs, this effect is enhanced in the sample #6 with a higher density of small dots; the pole diagram of sample #2 has been shown elsewhere [13] with no sign of such effect. Since it has increased with the density of QDs and not with their size,

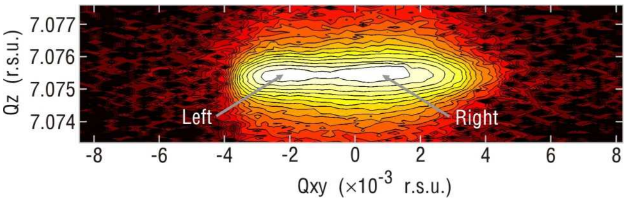

Figure 3 Reciprocal space map of the GaAs 004 reflection carried out on sample with exposed QDs ($\tau = 0.09\,$ML/s) and at the azimuth $\varphi = 84.3°$, same of Fig. 2(a). In this case, the distance between the left and right intensity maxima $\Delta Q_{xy} \simeq 2.55(\pm 0.15) \times 10^{-3}\,\mathrm{nm}^{-1}$, divided by $Q_z = 7.0754\,\mathrm{nm}^{-1}$ provides $\delta = 0.021°$ as the misorientation angle of the diffracting reciprocal vector. Experimental setup: CuKα_1 in triple-axis goniometry. r.s.u. = nm^{-1}.

the strain modulation should have its roots at the highly strained areas around the QDs, while the lattice below the dots is under expansive vertical strain [11], as illustrated in Fig. 2(c). After capping, the misorientation generated at the areas in-between the dots disappears given rise to another misorientation effect that has a different behavior. This latter is observed to be enhanced in the samples with larger QDs, Figs. 1(b-f). In this case, the probable origin of the weak strain field deeper in the substrate lattice is therefore the areas below the dots, which is now under compressive vertical strain [11]. For samples with QDs growth rates below $\tau = 0.01\,$ML/s, the direction of maximum misorientation of the 002 planes are observed to lay somewhere in-between [100] and [110] types of in-plane directions, Figs. 1(a-c). Above this value of growth rate, [100] is clearly the preferential misorientation direction. The reason might be related to changes in the shape of the QDs from domes to pyramid with [101] facets.

4 Conclusion We have demonstrated the sensitivity of the 002 reflection for studying the growth and capping of InAs/GaAs QDs system. It is capable of detecting tiny elastic tilt of the substrate lattice due to a long-range weak strain field modulation generated at the sample surface under the QDs. Changes in the azimuthal patterns of lattice tilt have be correlated to the QDs density, size, and shape. Fact that opens opportunity for using tube source diffractometer in systematic investigation of QDs based devices.

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References

[1] D. Bimberg, M. Grundmann, and N. N. Ledentsov, Quantum Dot Heterostructures (JohnWiley & Sons, Chicheste, 1999).
[2] Q. Xie, P. Chen, and A. Madhukar, Appl. Phys. Lett. **65**, 2051 (1994).
[3] D. Leonard, K. Pond, and P. M. Petroff, Phys. Rev. B **50**, 11687 (1994).
[4] I. Mukhametzhanov, Z. Wei, R. Heitz, and A. Madhukar, Appl. Phys. Lett. **75**, 85 (1999).
[5] I. Kegel, T. H. Metzger, A. Lorke, J. Peis, J. Stangl, G. Bauer, J. M. García, and P. M. Petroff, Phys. Rev. Lett. **85**, 1694 (2000).
[6] Q. Gong, P. Offermans, R. Nötzel, P. M. Koenraad, and J. H. Wolter, Appl. Phys. Lett. **85**, 5697 (2004).
[7] R. Otto, H. Kirmse, I. Häusler, W. Neumann, A. Rosenauer, D. Bimberg, and L. Müller-Kirsch, Appl. Phys. Lett. **85**, 4908 (2004).
[8] P. Offermans, P. M. Koenraad, J. H. Wolter, D. Granados, J. M. García, V. M. Fomin, V. N. Gladilin, and J. T. Devreese, Appl. Phys. Lett. **87**, 131902 (2005).
[9] G. Costantini, A. Rastelli, C. Manzano, P. Acosta-Diaz, R. Songmuang, G. Katsaros, O. G. Schmidt, and K. Kern, Phys. Rev. Lett. **96**, 226106 (2006).
[10] H. C. Chung, Y. F. Lai, C. P. Liu, Y. L. Lai, Y. C. Fang, and L. Hsu, Appl. Phys. Lett. **92**, 051903 (2008).
[11] M. Yang, S. J. Xu, and J. Wang, Appl. Phys. Lett. **92**, 083112 (2008).
[12] S. Tonomura and K. Yamaguchia, J. Appl. Phys. **104**, 054909 (2008).
[13] R. O. Freitas, T. E. Lamas, A. A. Quivy, and S. L. Morelhão, Phys. Stat. Sol. (a) **204**, 2548 (2007).
[14] W. Rudno-Rudziński, G. Sȩk, J. Misiewicz, T. E. Lamas, and A. A. Quivy, J. Appl. Phys. **101**, 073518 (2007).
[15] A. Authier, Dynamical Theory of X-Ray Diffraction (Oxford Univ. Press, Oxford, 2003).