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Microroughness and exciton localization in (Al,Ga)As/GaAs quantum wells

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We report on microscopic photoluminescence and photoluminescence excitation of thin $Al_{0.3}Ga_{0.7}As/GaAs$ quantum wells grown on exactly oriented (001) GaAs substrates. The experiments are done at low temperature by selectively exciting a few μm^2 of the sample with a low excitation intensity. The photoluminescence spectrum performed under these conditions shows sharp peaks appearing on the low-energy side of the main line of about 0.1-meV width. These features are attributed to localized exciton states at fluctuations of the effective quantum well thickness. On the other hand, scanning tunneling microscopy performed on 2×4 reconstructed GaAs (001) surfaces clearly evidences the presence of one-monolayer-deep elongated ''holes'' regularly oriented along the [110] direction with an average width of 6 nm. The same features are observed on 2×4 reconstructed $Al_{0.3}Ga_{0.7}As$ surfaces (on which quantum wells are grown), though the width regularity is less pronounced. This strongly suggests that excitons at low temperature are localized in such boxes. We also discuss the very different results obtained using both techniques (micro-photoluminescence and scanning tunneling microscopy) on quantum wells grown on vicinal (001) substrates. [S0163-1829(97)03708-9]

I. INTRODUCTION

Ultimate interface smoothness in semiconductor heterojunctions has been the object of long and numerous investigations. Interface roughness is detrimental in order to obtain sharp and controlled optical transitions in quantum wells (QW) and superlattices, and also high-mobility modulation-doped heterostructures.

We deal here with the topical case of (Al,Ga)As/GaAs quantum wells. The trapping of excitons onto interface defects, and the related Stokes shift, was first addressed by Weisbuch *et al.*[1] Progress in crystal growth, and in particular the optimization of growth interruptions at interfaces, has allowed the observation of resolved QW transitions corresponding to fluctuations of ±1 GaAs monolayer (ML) in QW thickness.[2–5] This could indicate that terraces atomically flat on a scale much larger than the exciton Bohr radius or the exciton diffusion length can be grown.

As the exciton is the local probe for the interface microscopic structure, by comparing the exciton diameter D_{ex} to the lateral size of the interface defect, L, a classification of the interface roughness can be obtained and is usually adopted in the literature.[6,7] The interface is called *pseudosmooth* if L is smaller than D_{ex}, then the exciton is sensitive to a mean QW width. The photoluminescence (PL) line is unique and the broadening is related to the QW width distribution. When L is larger than D_{ex}, the interface is called *smooth* and we can observe multiple PL lines which are very narrow and correspond to QW widths differing by one monolayer. Finally when L is of the same magnitude as D_{ex}, then the interface is *rough* and excitons can be localized.

It is nowadays considered that the (Al,Ga)As/GaAs interface has a bimodal roughness spectrum in Fourier space, i.e., that they consist of large atomically flat terraces on which a microroughness is superimposed.[8,9] This assumption comes from the observation of the 1-ML splitting measured in QWs grown with growth interruptions which is not constant across the sample,[8] from the study of Stokes shift and of excitonic thermal diffusion[9] but also from the observation of sharp individual lines in the photoluminescence or PL excitation (PLE) spectra of QWs,[10–14] either using micro-PL (Refs. 10–13) or near field scanning optical microscopy.[14] Indeed, though classical PL experiments have already given important information about the heterointerface structure, this technique shows its limitations arising from the spatial averaging of the spectral information. By performing ''microscopic'' spectroscopy we can get further insight into the interface structure. The high spatial resolution coupled to the spectral resolution and to a high quantum emission efficiency yields spectral information about lower-dimension structures.

This work presents results of micro-PL and micro-PLE studies of $(Al_{0.3}Ga_{0.7})As/GaAs$ QWs with various thicknesses grown by molecular beam epitaxy (MBE). The observation of sharp PL lines in the low-energy tail of the QW PL spectra is correlated to a study by scanning tunneling microscopy (STM) of GaAs and $Al_{0.3}Ga_{0.7}As$ (001) 2×4 reconstructed surfaces, grown in the same MBE machine under similar conditions, and exhibiting the systematic appearance of 1-ML-deep holes of width comparable to the exciton Bohr radius, able to localize excitons with the suitable energy.

Furthermore, our results are compared to those obtained with both characterization techniques on substrates slightly misoriented (2° and 4°) towards (111) Ga, where the presence of 1-ML step array totally modifies the situation with regard to microroughness as well as optical properties.

II. EXPERIMENTAL DETAILS

Experiments were carried out in a MBE system equipped with a standard reflection high-energy electron diffraction

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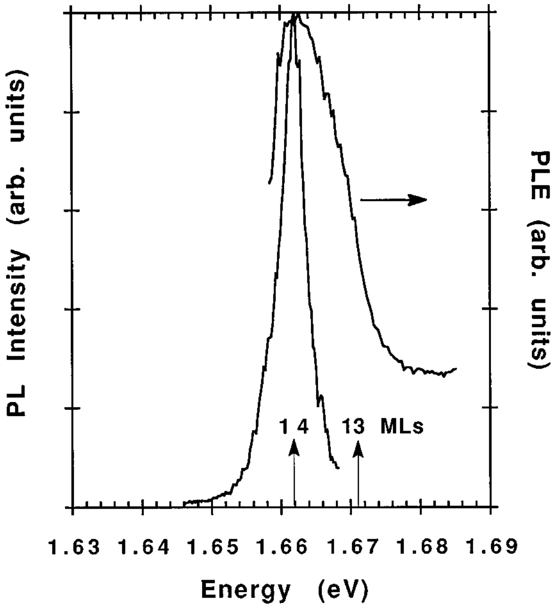

FIG. 1. PL and PLE spectra of the 14-ML nominal QW. The PL excitation energy is 2.54 eV and the PLE detection is set at 1.657 eV. The power intensity is 200 μW. The arrows indicate the position of the excitonic transitions in a 14- and 13-ML-wide QW.

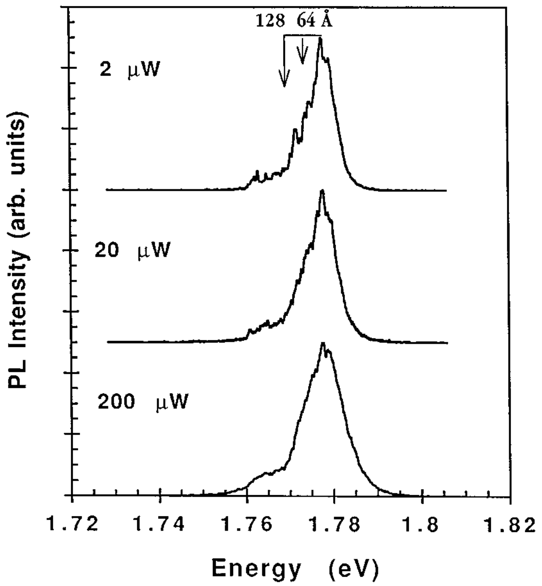

FIG. 2. PL spectrum of the 7-ML nominal QW as a function of the excitation power. Arrows indicate the localization energies of excitons confined in boxes with 64- and 128-Å diameter (see text).

(RHEED) facility and coupled to a STM chamber. The samples studied were grown at 600 °C on both nominal and vicinal [4° off towards (111) Ga] surfaces by using a rotating substrate holder. The heterostuctures studied in PL were three QWs of 7, 14, and 28 ML separated by 500 Å of $Al_{0.3}Ga_{0.7}As$ barriers. Growth interruptions optimized by the RHEED intensity recovering procedure were performed on each interface in order to improve their smoothness.[7] The samples for STM measurements were quenched below 450 °C within 30 s. The STM filled state images were taken with sample bias voltages between -4 and -2 V and a tunneling current range of 0.05–0.5 nA.

PL and PLE spectra have been recorded at 4 K using an Ar-pumped dye laser and a tunable Ti-sapphire laser as the excitation sources. The sample is mounted on the cold finger of a helium cryostat. The laser beam is focused on the sample by a microscope objective with a large numerical aperture (0.6). The spot diameter is about 1 μm. The laser power is usually a few hundred μW and can be reduced to 100 nW. The PL signal is analyzed in a 1-m grating monochromator and detected by a cooled GaAs photomultiplier using a conventional photon counting system. An intensified Si-diode multichannel analyzer was also used for detection. The optimal spectral resolution obtained was 0.05 meV.

III. EXPERIMENTAL RESULTS

A. Nominal sample

We have performed PL and PLE of all QWs at different excitation power densities at low temperature. In Fig. 1 are shown PL and PLE spectra of the 14-ML nominal QW. Since there is no Stokes shift between the PL line and the PLE spectrum, we can consider that the QW interfaces are well smoothed after growth interruptions. The PLE line is twice as wide as the PL line, which indicates that at least two terraces were formed during the growth, corresponding to 13- and 14-ML-wide wells. The energy difference corresponds to the difference in confinement energy between the two QWs. It is worth noticing that the signal-to-noise ratio is 10^3 and the structures appearing on both PL and PLE spectra are characteristic of the 1-μm^2 scanned sample area. The same signal-to-noise ratio is valid for all our PL and PLE experiments.

Figure 2 represents the PL spectrum of the 7-ML nominal QW as a function of excitation power. We clearly see that as the power density is decreased sharp peaks appear only on the low-energy side of the PL line. These sharp features are completely reproducible from scan to scan at the same position and their intensity is much larger than the magnitude of noise. They are attributed to localized exciton states on growth islands present at the QW interface. As can be expected, these sharp structures are more pronounced when the QW width is narrow, since in this case the exciton is very sensitive to the microscopic structure on the interface. This can be seen in Fig. 3 where the PL spectra of the two nominal QWs (7 and 14 ML) at low excitation power are represented. From the above observations we may conclude that although the interfaces after growth interruption are smoothed, excitons can be localized on islands of different sizes at the heterointerface. This result suggests that roughness on a smaller scale is superimposed on the wide terraces initially formed.

Let us examine now the STM of 2×4 reconstructed GaAs surfaces after rapid cooling of the samples. Figure 4 (large scan area: 400×400 nm^2) reveals a growth front which consists of three levels with terraces elongated along the [110] direction. This result is in agreement with previous reports for similar growth conditions[15,16] and with our PLE observation of at least two QW widths. Actually, looking closer at these wide terraces, they clearly appear to be textured by one-monolayer-deep holes of relatively homogeneous distribution and size. They are elongated along [110] and their

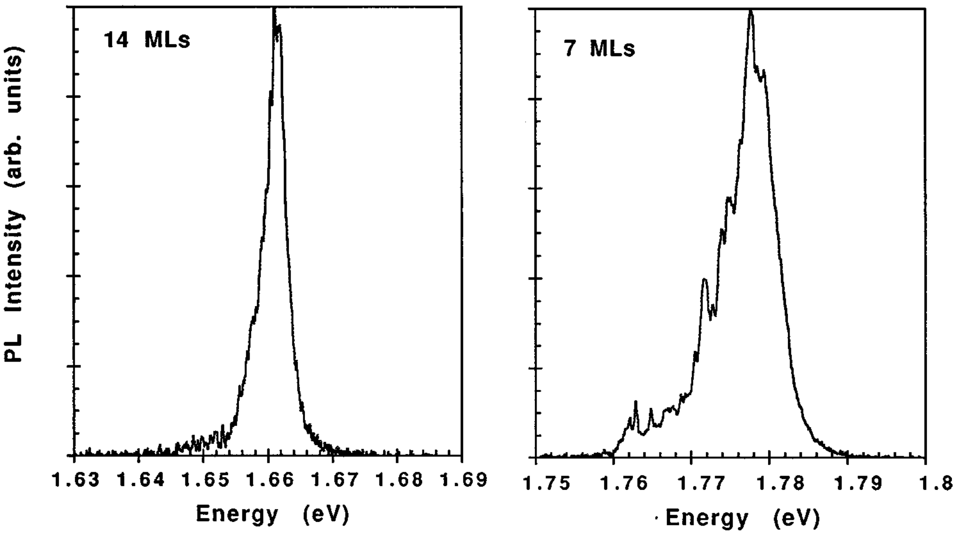

FIG. 3. PL spectra of two nominal QWs (2 μW excitation power).

widths along [110] are most frequently four 2×4 blocks, i.e., 64 Å. A study of previously published STM images of III-V arsenide surfaces shows that such holes are present on 2×4 reconstructed surfaces whether the material is GaAs (Refs. 15–20) or $Ga_{1-x}In_xAs$.[21] The STM observation reported in Fig. 5 of a 2×4 reconstructed $Al_{0.3}Ga_{0.7}As$ surface (100 ×100 nm^2 scan) is even more relevant for the correlation between micro-PL and STM studies. This is indeed the surface on which QWs are grown. We see in Fig. 5 that the same microroughness is observed, i.e., that the four (2×4)-block-wide and one-monolayer-deep holes still exist.

We have also performed resonant PL experiments by varying the excitation wavelength close to the QW excitonic transition. The sharp peaks then change slightly in position depending on the excitation wavelength. This means that we are able to excite selectively fundamental or excited states of an island with a certain size. However, we observe that the number of peaks does not depend either on the excitation wavelength or on the position on the sample. This proves that in this sample the islands have a given size and this result is in agreement with the STM images. The arrows in Fig. 2 indicate the localization energies (4 and 10 meV, respectively) of excitons confined in 1-ML-deep boxes with 64 and 128 Å diameter, showing that the energy range spanned by the sharp peaks may easily correspond to exciton states localized in such boxes.

Our results are comparable with the microroughness scale reported in the literature. The average localization energy measured in a 6-ML-wide well is 4 meV (corresponding to about 5-nm scale),[9] in agreement with our result. A lateral roughness scale of ~100 Å (with no precise orientation) explains the electron mobility limitation at low temperature in high-quality QW.[22] A roughness scale of 30–100 Å has frequently been quoted from PL linewidths and Stokes shifts.[7,14,23,24]

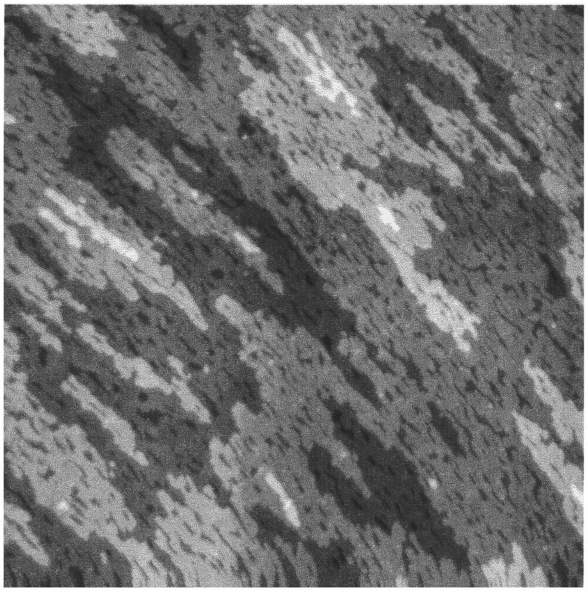

FIG. 4. Large area (400×400 nm^2) STM scan (filled states) of the as-grown GaAs (001) surface. Each gray level corresponds to 1 ML height (1 ML=2.83 Å).

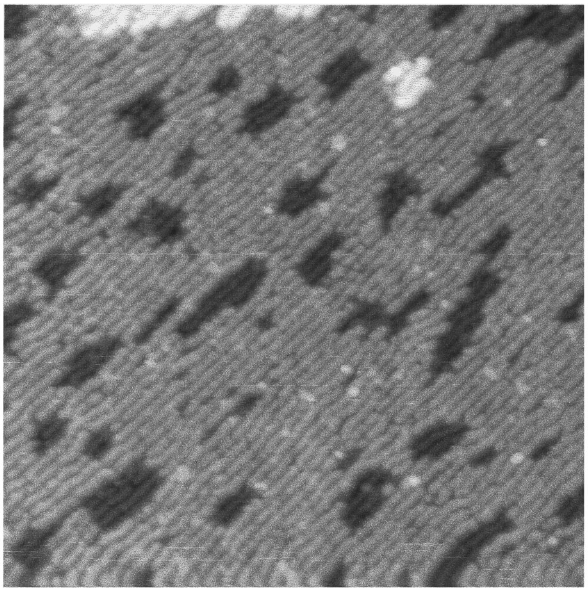

FIG. 5. STM image (100×100 nm^2) of a 2×4 reconstructed $Al_{0.3}Ga_{0.7}As$ surface.

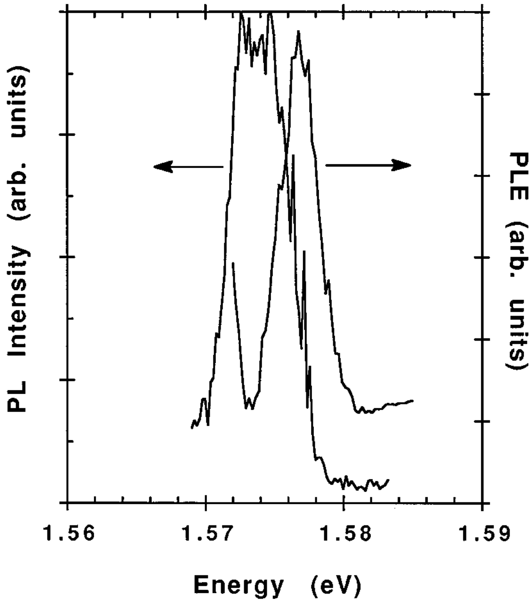

FIG. 6. PL and PLE spectra of the 28-ML-wide vicinal *A* QW (150 nW excitation power).

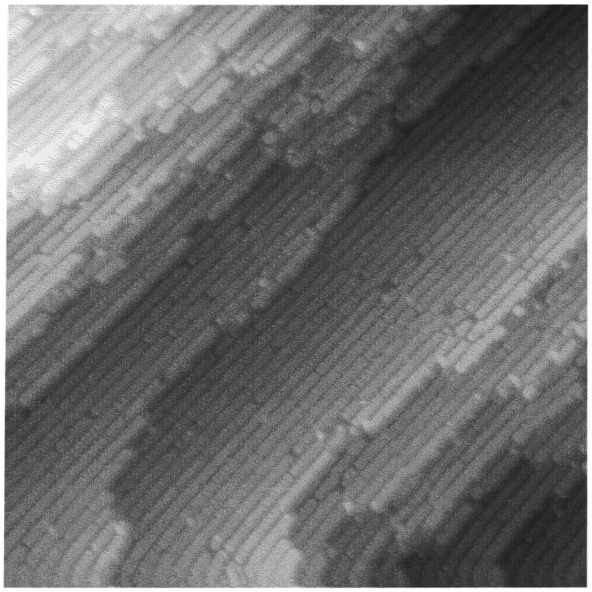

FIG. 7. STM image of a 2° misoriented (111)Ga surface (75×75 nm^2).

B. Vicinal sample

The early motivation of growing and studying heterostructures on vicinal substrates was the possibility to build new low-dimensional structures such as quantum wires[25] or lateral superlattices.[26] In this work, we shall study vicinal surfaces in order to compare our previous results on nominal (001) surfaces, with another rough system. Ideally, a vicinal surface is formed by staircaselike terraces regularly distributed. The studied samples are 4° cutoff GaAs (001) substrates towards Ga (111) (vicinal *A*) that produce misorientation imposing monolayer steps with a mean terrace length of 40 Å. The average terrace width is determined by the disorientation angle of the surface referring to the (001) plane of GaAs, considering a step of one-monolayer height. We shall not discuss in this work the difficult case of surfaces disoriented towards (111) As.[7,12]

Figure 6 shows PL and PLE spectra of the 28-ML-wide vicinal QW excited with 150 nW power intensity. We observe that the PLE spectrum is blueshifted by 2 meV as compared to the nominal sample. This effect has already been reported in the literature[27] and was attributed to the additional quantum confinement effect due to the lateral modulation of the potential. A second observation is the Stokes shift between PL and PLE as compared to the nominal sample. In the case of a 28-ML-wide well the Stokes shift is 3 meV and increases as the well width decreases. This suggests that important localization phenomena can occur in the vicinal sample. Finally, very sharp peaks appear on both sides of the PL and PLE spectra, suggesting again localized exciton states. However, when we perform resonant PL spectra as in the nominal sample, the localization sites depend strongly on the position at the interface. Thus we may conclude that in contrast to the nominal case a different kind of roughness exists in the vicinal sample.

The STM image of a 2° misoriented towards (111) Ga surface is shown in Fig. 7 (let us remember that the spectra in Fig. 6 correspond to 4°, i.e., 40-Å average terraces). Two salient features appear in Fig. 7. First, although the terrace width has the right mean periodicity a strong disorder appears and second the holes observed on nominal surfaces are absent.

In light of the STM study, the micro-PL and micro-PLE results can be understood by considering a QW grown on a (111) Ga misoriented surface as an array of disordered wires or boxes. Isolated wires may be excited separately but excitons are allowed to diffuse towards a neighboring wider one (perpendicular or along the steps) before radiative recombination. This may explain the large Stokes shift measured and the appearance of the sharp transitions on the low- and high-energy sides of the PL and PLE lines. Note that these peaks depend strongly on the position on the sample and that in contrast to the nominal case, the roughness associated with the previously described holes is not observed.

IV. DISCUSSION

The origin of the holes appearing on the 2×4 (001) surface is worth discussing. They could have a purely stochastic origin and be due to the two-dimensional nucleation growth and the coalescence of laterally growing islands. However, the homogeneity in size, in particular along [110], is somewhat surprising for such a stochastic phenomenon. Figure 8 shows a 200×200 nm^2 STM image of a 2×4 GaAs nominal surface which was annealed at 600 °C for 45 min under As$_4$ (beam pressure of 3×10^{-6} Torr). Actually, the holes are still present with a very regular distribution. This suggests that such a situation is an equilibrium one coming from the self-compensated electronic nature of the 2×4 unit cell which is the building block of the surface.[17,28] The role of the 2×4 reconstruction on the vacancy formation is confirmed by recent experiments showing that the adsorption of a monolayer fraction of tellurium on the GaAs(001) inhibits the 2×4 reconstruction and consequently the holes.[29] On the other hand, the holes are not observed on vicinal surfaces. One possible reason is that since the growth on such a vicinal

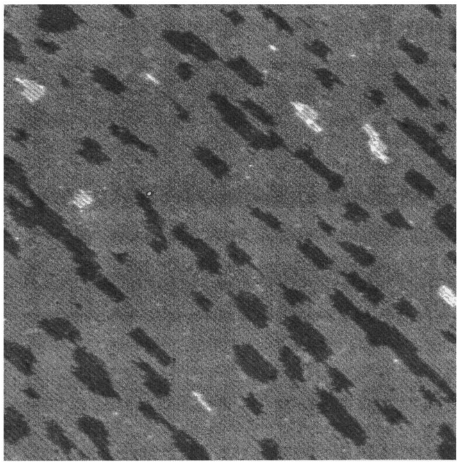

FIG. 8. Large area (200×200 nm^2) STM scan of a 2×4 reconstructed GaAs (001) surface obtained after the annealing of a buffer layer for 45 min at 600 °C.

surface follows a step-flow mode, two-dimensional islands are not formed and therefore hole formation by island coalescence cannot occur. This would mean again that the presence of holes corresponds to the equilibrium state of a 2×4 reconstructed nominal surface. Finally, it is worth recalling that other reconstructions of the GaAs(001) surface exist, such as the 3×1 or the $c(4\times4)$ in which such holes are not observed.[30]

Another interesting point should be mentioned concerning the relevance of the micro-PL experiments. The density of holes observed in STM is estimated to be 10^3 per μm^2, the holes having relatively homogeneous width with a few percent of size dispersion. Taking also into account that the excitation mean power intensity is 1 μW, it is clear that localized exciton states are not saturated and can be observed. By considering also that the full width at half maximum (FWHM) of the PL line is 10 meV and the resolved width of the sharp peaks is 0.1 meV (Fig. 2), then the observation of such sharp features is only possible under micro-PL experimental conditions. Indeed, if the size of the laser spot is increased then the number of scanned holes will proportionally increase and the structures will progressively disappear because of inhomogeneous broadening.

V. CONCLUSION

The combined analysis of optical measurements at a micrometer scale and STM observations has allowed further insight into the phenomenon of exciton localization in $Al_{0.3}Ga_{0.7}As$/GaAs QWs grown on nominal or vicinal substrates. The results show that on large terraces a microroughness is superimposed, able to localize excitons at low temperature. The microroughness is due to one-monolayer-deep holes with a given width along the [110] axis of four 2×4 blocks and these holes are observed on GaAs as well as on $Al_{0.3}Ga_{0.7}As$ surfaces. This image of a bimodal roughness scale is not valid in the case of QWs grown on substrates misoriented towards (111)Ga.

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