A growth method to obtain flat and relaxed $In_{0.2}Ga_{0.8}As$ on GaAs(001) developed through *in situ* monitoring of surface topography and stress evolution

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Abstract

In this paper we develop a growth process for obtaining flat and relaxed $In_{0.2}Ga_{0.8}As$ layers on GaAs (001). The process designed is based on the results obtained by *in situ* and real time characterization of surface morphology and layer relaxation. In particular our results show that for growth temperatures $T_s \leq 200$ ºC, the relaxation of $In_{0.2}Ga_{0.8}As$ layers is inhibited and the morphology does not evolve to a crosshatched pattern. After growth thermal treatments of these low temperature (LT) $In_{0.2}Ga_{0.8}As$ layers induce the development of a very faint (rms = 0.5 nm) crosshatched-like morphology. The relaxation process during the thermal annealing is strongly asymmetric and the layers present a high final strain state. By growing on top of the LT layer another $In_{0.2}Ga_{0.8}As$ layer at higher temperature, relaxation is increased up to $R \approx 70\%$ and becomes symmetric. Depending on the growth process of the top layers morphology evolution differs, resulting better morphologies for top layers grown by atomic layer molecular beam epitaxy (ALMBE) at $T_s = 400$ ºC. We have obtained 400 nm $In_{0.2}Ga_{0.8}As$ layers with a final degree of relaxation R = 70% and very flat surfaces (rms = 0.9 nm).

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The growth of good quality relaxed buffer layers on III-V semiconductor substrates would constitute a great advance for obtaining high performance devices as designed by band gap engineering and without restrictions in the lattice parameter. However, even for low mismatched layers (< 2%), the growth front usually develops a crosshatched morphology that constitutes a substantial limitation for subsequent device fabrication [1,2].

The evolution of these crosshatched morphologies is closely related both to the growth kinetics and the relaxation processes. In order to study in depth these two processes we have performed real time measurements during the growth of $In_{0.2}Ga_{0.8}As$/GaAs(001) under very different conditions for optimising the morphology and at the same time achieving a high final degree of relaxation of the mismatched layer. The surface morphology evolution has been assessed by means of in-situ laser light scattering (LLS) [3,4] and the relaxation process is followed by stress measurements based on the deflection of a laser beam on a cantilever shaped substrate [5].

Our results allow to develop growth processes that improve morphology of highly relaxed layers. As a test we have grown a 400nm thick $In_{0.2}Ga_{0.8}As$ layer with relaxation R ≈ 70% and roughness rms = 0.9, very flat surfaces if compared with 400nm thick $In_{0.2}Ga_{0.8}As$ layers grown by a conventional molecular beam epitaxy (MBE) process (rms = 4.7 nm, R = 73%).

The samples studied consist of nominal $In_{0.2}Ga_{0.8}As$ layers grown at 1 monolayer per second (Ml/s) on on-axis GaAs (001) substrates with an average etch pit density of 10^4 cm^{-2}. After 100/200 nm thick GaAs buffer layers grown at a substrate temperature T_s=580 ºC, $In_{0.2}Ga_{0.8}As$ layers were either grown by conventional MBE at T_s=500ºC or by atomic layer molecular beam epitaxy (ALMBE) [6] at T_s= 400ºC and T_s= 200 ºC . Arsenic pulse (flux and duration) in ALMBE growth was established by *in situ* reflectance difference (RD) characterisation [7].

Surface morphology evolution has been monitored during growth by means of *in situ* real time laser light scattering measurements. The sample surface was illuminated with a 10 mW He-Ne laser (λ=633 nm) at an angle of incidence θ_i= 50º, and the light scattered at an angle θ_s= 0º was detected with a silicon photodiode [4]. In order to follow the crosshatched morphology evolution, where roughness mainly develops along <110> directions, we carefully align before growth the [110] direction of the sample into the light scattering detection plane. This geometry allows us to monitor in real-time the evolution of the roughness along [110] direction, with maximum sensitivity for lateral length scales on the surface plane of 800 nm [8]. We can also rotate the sample during growth to obtain information about the surface roughness along other directions. Final surface morphology characterization has been completed by *ex situ* atomic force microscopy (AFM).

The relaxation process has been followed during growth by *in situ* and real time strain measurements based on the determination of the stress induced substrate curvature through the deflection of a laser beam [5, 9]. For this purpose, the 350 μm thick GaAs (001) substrates were cut along [110] or $[1\bar{1}0]$ directions, shaped as cantilever, and mounted on a special substrate holder that lets the sample bend. The stress in the layer is related to the substrate curvature by Stoney’s equation [9]. Composition and final strain of the $In_{0.2}Ga_{0.8}As$ layers were obtained by X-ray diffraction measurements of (004) and (115) reflections, taken in both <110> directions.

Crosshatched morphology has been related [3, 10, 11] to variations in surface diffusion due to the presence at the surface of inhomogeneous stress fields associated with misfit dislocations in the layer. The reduction of the growth temperature in order to inhibit the surface diffusion has been proved as a good way to improve the flatness of the layers [12].

To find a temperature at which morphology does not evolve at all we have grown $In_{0.2}Ga_{0.8}As$ layers at different substrate temperatures while following the morphology evolution with the *in situ* LLS technique. For temperatures above 200 ºC, the LLS signal always increased, indicating the development of surface roughness. However, for $T_s \leq 200$ºC the LLS signal did not increase even for layers as thick as 300 nm. Unfortunately, X-ray results show that these layers are fully strained (R=0%) indicating that at 200ºC the relaxation process is inhibited.

In order to relax these layers, thermal treatments were applied in the MBE chamber directly after growth. Morphology and relaxation evolution were *in situ* monitored both during growth and subsequent thermal treatment. We have observed that within the accuracy of our *in situ* measurements all layers remain coherent to the substrate during the ALMBE growth at T_s = 200ºC (notice that our experimental set up allow to determine the layer indium composition with an error of 0.5% as confirmed by *ex situ* X-ray diffraction measurements). Figure 1 shows the real time relaxation measurement during thermal annealing of 200 nm thick $In_{0.2}Ga_{0.8}As$ layers grown at T_s = 200ºC taken along both $\langle 110 \rangle$ directions. Immediately after layer growth, samples were cooled down, and then heated as fast as possible up to 500 ºC. Arsenic cell was opened when the RHEED pattern showed a 4x2 reconstruction (T_s = 480ºC). The layers remain at T_s = 500 ºC during the same time period as if the layer would have been grown at high temperature.

Our results (fig.1) show that relaxation along [110] direction takes place very quickly, once 200ºC have been surpassed, and remains constant at a temperature around 500ºC. By contrast, along $[1\bar{1}0]$ direction different processes seem to be activated with increasing temperature. Above 200ºC a small and slow relaxation occurs, which continues until approximately 390 ºC. From this temperature up to 480ºC we find a plateau in relaxation, and above 480 ºC another relaxation process becomes active, which keeps on working at 500ºC. After 5 minutes at this temperature the relieved strain remains constant. We thus obtain that relaxation is clearly asymmetric, with R=32 % and R=27 % along [110] and $[1\bar{1}0]$, respectively. Furthermore, the relaxation degree of these layers is much lower than that obtained for similar layers grown by conventional MBE, even though they have stayed the same time at high temperature. This difference could be due to the existence of only one surface (instead of fresh surfaces continuously provided during growth) as supplier of nucleation sites for dislocation formation during thermal treatment.

Besides relaxation of the growing $In_{0.2}Ga_{0.8}As$ layers we have also studied their morphology evolution by *in situ* LLS. Figure 2 shows the *in situ* LLS signal evolution during thermal treatment of a 300 nm thick $In_{0.2}Ga_{0.8}As$ layer grown by ALMBE at T_s = 200ºC. AFM images of similar layers before and after the thermal cycle are shown.

Although *in situ* morphology measurements do not detect roughness development during growth, post growth AFM characterization (bottom of fig. 2) reveals the presence of ripples along [110] direction. These ripples, with approximate heights of 4.6 nm, show a clear periodicity of 42 nm, far away from the lateral length scales detectable with our LLS experimental set-up (maximum sensitivity for 800 nm), which explains why we do not detect their formation during growth. The presence of these ripples points out that surface diffusion is not completely inhibited at T_s = 200 ºC, and that in our growth conditions, [110] direction could be the preferential surface diffusion direction [13].

Otherwise, we find that when the substrate temperature is increased, the LLS signal takes off abruptly, indicating that the morphology starts to evolve towards features that now are detectable by our LLS experimental set-up. Discrete points on fig. 2 correspond to LLS signal when [100] and $[1\bar{1}0]$ directions are contained in the scattering plane, showing that LLS signal is mainly concentrated in <110> directions [8, 14]. From these results, we can establish that during thermal annealing, while growth is stopped, a crosshatched morphology is developed.

The final morphology after the thermal treatment is shown in AFM image at the top of fig. 2. It shows a very flat crosshatched surface (rms= 0.5 nm) where large terraces of monoatomic step height are clearly resolved.

In summary, layers grown at T_s = 200 ºC and subjected to thermal treatments relax about 30 % of the misfit strain and developed a faint crosshatched morphology, but the very flat final surface (rms = 0.5 nm) make them promisingly suitable for subsequent growth. So, in order to improve the relaxation degree of these very smooth samples we grew 200 nm thick $In_{0.2}Ga_{0.8}As$ layers by MBE at T_s = 500 ºC or by ALMBE at T_s = 400 ºC on top of the thermal annealed 200 nm thick $In_{0.2}Ga_{0.8}As$ epitaxies grown at T_s = 200 ºC.

The results obtained from the *in situ* recorded relaxation behaviour during the whole growth process show that, despite the asymmetric relaxation behaviour during the thermal annealing (see fig. 1), the final degree of relaxation after growth of the top $In_{0.2}Ga_{0.8}As$ layer does not depend on the azimuth, getting values around 70%. Same behaviour is observed when top $In_{0.2}Ga_{0.8}As$ layers are grown by MBE at T_s = 500 ºC or by ALMBE at T_s = 400 ºC.

However the morphology evolution is strongly dependent on the top $In_{0.2}Ga_{0.8}As$ layers growth process. The *in situ* recorded LLS evolution during the whole growth process for samples with top $In_{0.2}Ga_{0.8}As$ layers grown by MBE at T_s = 500 ºC and ALMBE at T_s = 400 ºC is shown in fig. 4. We have also introduced the LLS signal evolution of 400nm $In_{0.2}Ga_{0.8}As$ layer grown by a conventional MBE process for comparison. AFM images of the final morphology of these layers are also shown.

We want to notice that the LLS signal values for the 200 nm thick low temperature grown $In_{0.2}Ga_{0.8}As$ layers with post-growth thermal treatment are much lower than the corresponding to the layer grown by a conventional MBE process at the same thickness. When growth goes on after thermal treatment, the LLS signal value increases with thickness, but always remains well below that corresponding to the $In_{0.2}Ga_{0.8}As$ sample grown by a standard MBE growth process. From AFM images, we have obtained roughness rms values of 4.7 nm for the $In_{0.2}Ga_{0.8}As$ layer grown by a standard MBE process, rms = 2.4 nm for the sample with top layer grown by conventional MBE and rms = 0.9 nm when the top layer was grown by ALMBE at T_s = 400 ºC.

Conclusions

By using *in situ* and real time characterization techniques we have obtained continuous information in each growth run about the morphology evolution and the relaxation process. In particular our results show that the relaxation of $In_{0.2}Ga_{0.8}As$ layers is inhibited at $T_s \leq$ 200 ºC, but surface diffusion is not completely suppressed. Thermal treatments of $In_{0.2}Ga_{0.8}As$ layers grown at T_s = 200 ºC induce the development of a very faint (rms = 0.5 nm) crosshatched-like morphology even when growth is stopped. The relaxation process during the thermal annealing is strongly asymmetric and the layers remain highly strained.

To increase the relaxation degree in the layers, a subsequent $In_{0.2}Ga_{0.8}As$ top layer has to be grown at higher temperature. With that procedure, the final strain state (R ≈ 70%) is independent on the azimuth and on the growth process (MBE at T_s = 500 ºC or ALMBE at T_s = 400 ºC). However, the morphology evolution depends on the growth process used in the top layer, resulting better morphologies when top layers are grown by ALMBE at T_s = 400 ºC.

In summary, we have developed a growth process that allows us to obtain 400 nm $In_{0.2}Ga_{0.8}As$ layers with a very smooth surface, rms = 0.9 nm, and a final degree of relaxation of R = 70%. These results reflect a clear improvement compared with same thickness $In_{0.2}Ga_{0.8}As$ layers obtained by a conventional MBE process (rms = 4.7 nm, R = 73%).

Acknowledgements

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Figure captions

Figure 1: *In situ* measured relaxation along $[1\bar{1}0]$ (closed circles) and [110] (open circles) directions during thermal annealing up to 500 ºC of a 200 nm thick $In_{0.2}Ga_{0.8}As$ layer grown by ALMBE at T_s = 200 ºC. The left part of the figure shows the relaxation evolution versus temperature until the sample reaches 500 ºC. In the right part, behaviour versus time, at a constant temperature, T_s = 500 ºC, is plotted.

Figure 2: *In situ* LLS signal evolution during thermal treatment of a 300 nm thick $In_{0.2}Ga_{0.8}As$ layer grown by ALMBE at T_s = 200ºC. Discrete points correspond to LLS signal when [100] (open circle) and $[1\bar{1}0]$(closed circle) directions are contained in the scattering plane. AFM images of similar layers before (at the bottom) and after (at the top) the thermal cycle are shown. Note the different scan area size in the AFM images.

Figure 3: *In situ* recorded LLS evolution during the whole growth process: 200 nm thick $In_{0.2}Ga_{0.8}As$ layer grown by ALMBE at T_s = 200ºC, thermal annealing of this layer and 200 nm thick $In_{0.2}Ga_{0.8}As$ layer grown by ALMBE at T_s = 400ºC or by MBE at T_s = 500 ºC. We have also introduced the LLS signal evolution of 400nm $In_{0.2}Ga_{0.8}As$ layer grown by a conventional MBE process for comparison. The detailed LLS evolution during the thermal treatment was previously shown in fig. 2. AFM images of the final morphology of these layers are also shown.

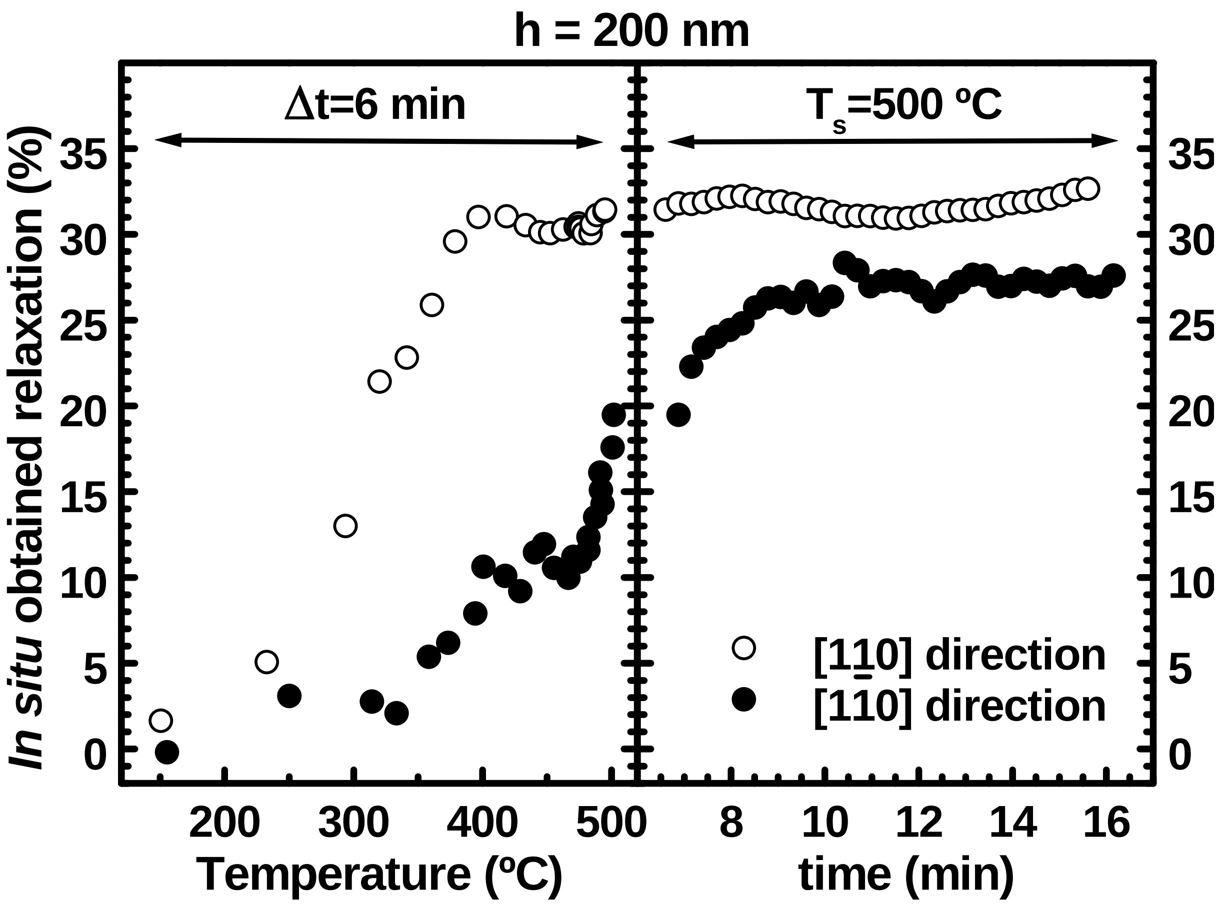

h = 200 nm
Δt=6 min
Ts=500 ºC
In situ obtained relaxation (%)
0
5
10
15
20
25
30
35
200
300
400
500
Temperature (ºC)
8
10
12
14
16
time (min)
[11̲0] direction
[110] direction

Fig.1

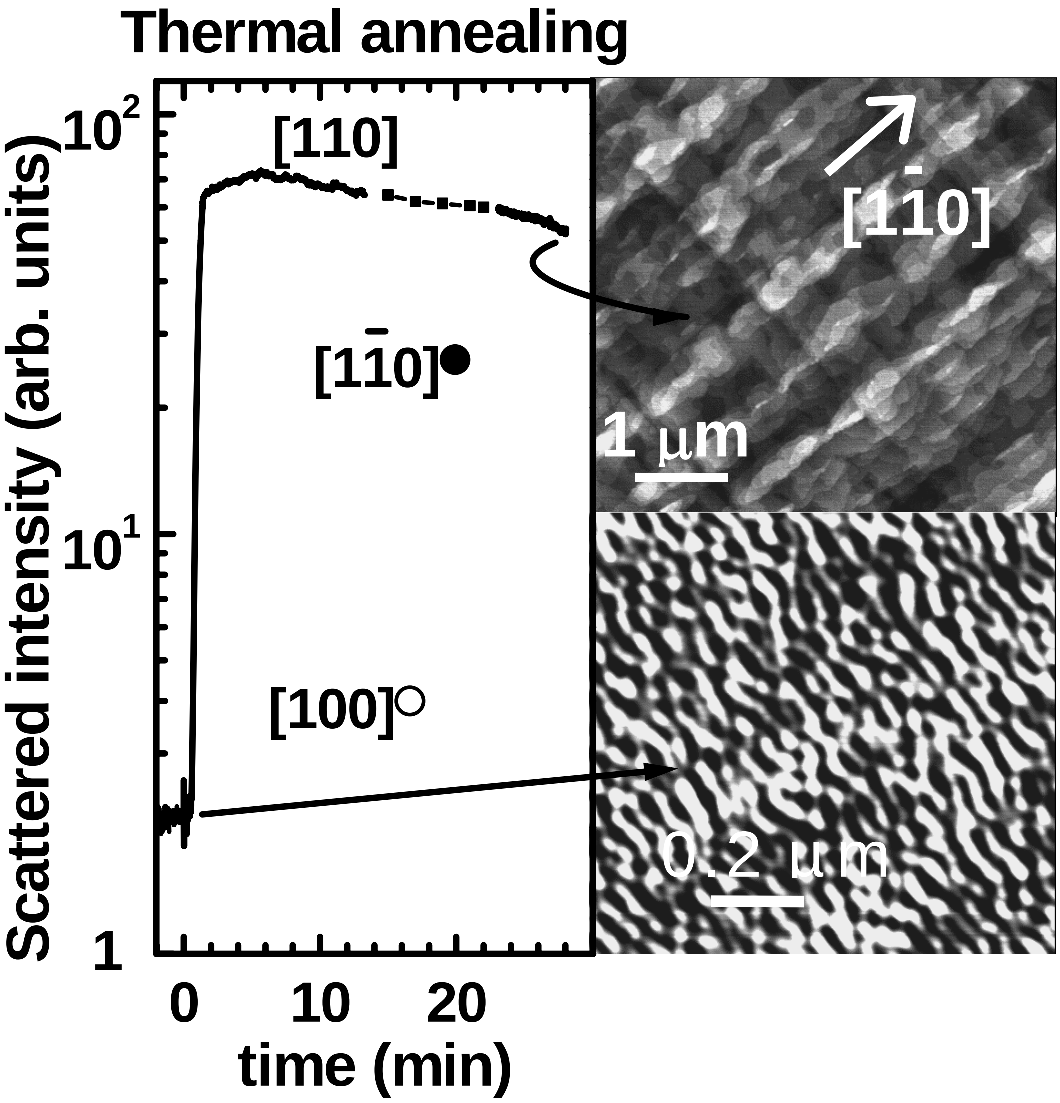
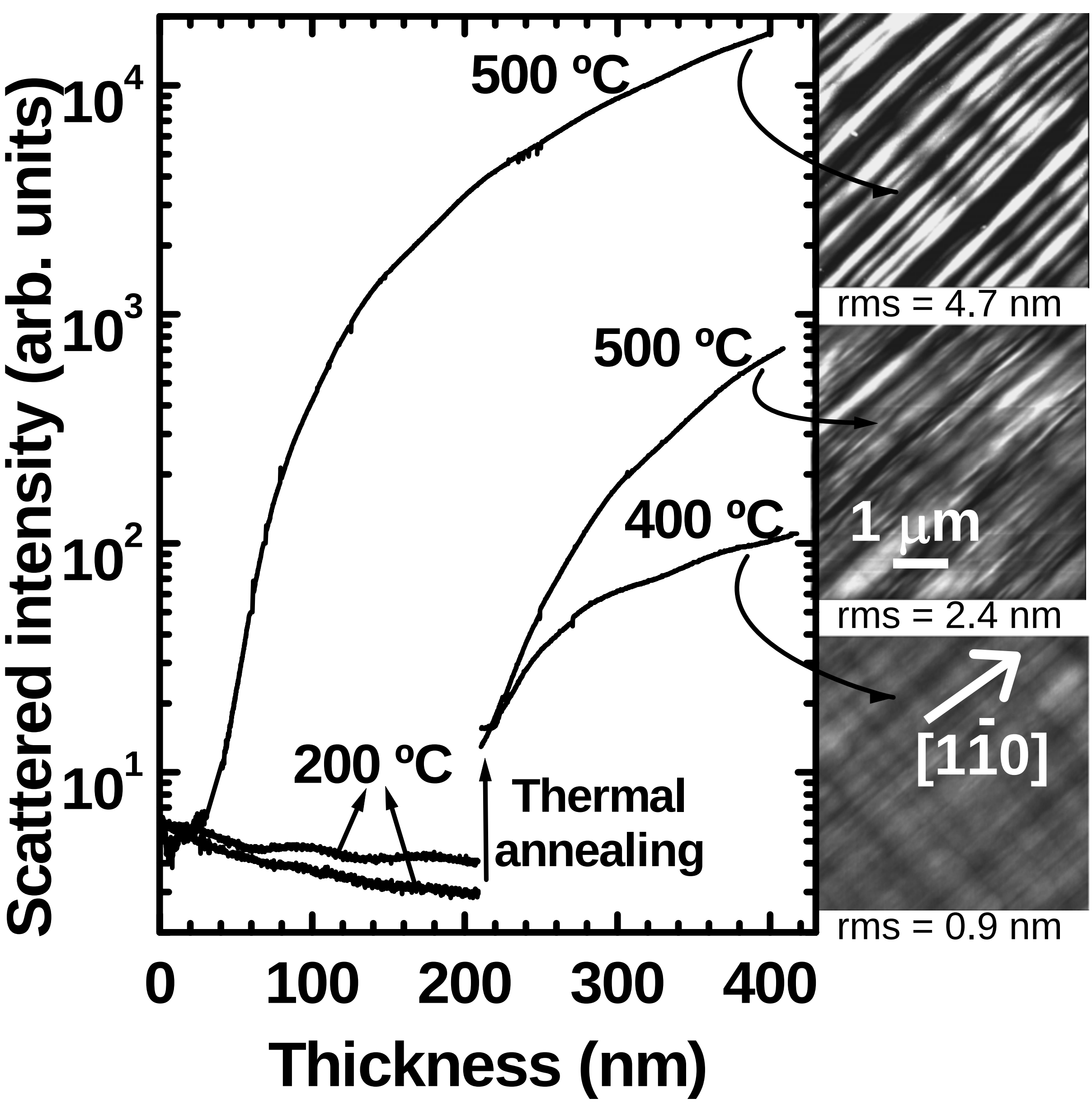

Thermal annealing
Scattered intensity (arb. units)
10^2
10^1
1
[110]
[1$\bar{1}$0]
[100]
0
10
20
time (min)
[1$\bar{1}$0]
1 μm
0.2 μm

Fig. 2.



500 ºC
500 ºC
400 ºC
200 ºC
Thermal annealing
Scattered intensity (arb. units)
Thickness (nm)
10^4
10^3
10^2
10^1
0
100
200
300
400
rms = 4.7 nm
rms = 2.4 nm
rms = 0.9 nm
1 μm
[1-10]

Fig. 3.