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X. Wallart, D. Deresmes, F. Mollot. Growth of strained Ga_{1-x}In_xP layers on GaP (001) by gas source molecular beam epitaxy: similarities and differences with the growth of strained arsenides. *Journal of Crystal Growth*, 2001, 227-228, pp.255-259. 10.1016/S0022-0248(01)00693-5 . hal-00152087

HAL Id: hal-00152087

<https://hal.science/hal-00152087>

Submitted on 19 Aug 2022

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Growth of strained $\text{Ga}_{1-x}\text{In}_x\text{P}$ layers on GaP (00 1) by gas source molecular beam epitaxy: similarities and differences with the growth of strained arsenides

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Using reflection high energy electron diffraction and atomic force microscopy, the growth of $\text{Ga}_{1-x}\text{In}_x\text{P}$ alloys on GaP (00 1) with x varying from 0.2 to 1 (InP) is investigated and compared to that of arsenides on GaAs (00 1) or InP (00 1). At 520°C, the evolution of the critical thickness for 3D growth versus In content is rather similar to that observed in the GaInAs/GaAs system. For $x \leq 0.5$, 3D growth leads to the development of wire-like structures along the [1 1 0] direction which can be related to recent results on the phosphide surface reconstructions. Finally, for the growth of InP on GaP at 520°C, the critical thickness is 2.1 MLs and we observe a small density of very large islands, in contrast to the InAs/GaAs case. At 400°C, the critical thickness decreases (1.7 MLs) as well as the island mean size whereas the density increases. We discuss this behavior in terms of surface energy.

Keywords: A1. Surfaces; A3. Molecular beam epitaxy; B1. Phosphides; B2. Semiconducting indium gallium phosphide

1. Introduction

Up to now, strained layer growth of III–V semiconductors has been mainly devoted to the GaInAs/GaAs and GaInAs/InP material systems. However, as an Al-free alloy with a rather large band gap, GaInP turns to be an interesting candidate for microwave as well as for optoelectronics applications. In most practical cases, band gap engineering purposes require the use of strained GaInP alloys but, it is not

straightforward that the knowledge got from the study of strained GaInAs growth can apply directly to GaInP alloys, considering for instance, the difference in surface reconstructions and hence energies as well as in mixing enthalpies for both alloys. Previous work on the growth of strained GaInP alloys have mainly focused on the growth on GaAs [1] or InP [2,3] substrates whereas a few studies have explored the growth of InP on GaP either by organometallic vapor-phase epitaxy [4,5] or by chemical beam epitaxy [6]. However, to the best of our knowledge, no systematic study on GaP substrates, which allows a direct comparison with the well-known GaInAs/GaAs system, has been undertaken.

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In this work, using reflection high energy electron diffraction (RHEED) and atomic force microscopy (AFM), we investigate the growth of $\text{Ga}_{1-x}\text{In}_x\text{P}$ alloys on GaP (001) substrates with the In composition x varying from 0.2 to 1.

2. Experimental procedure

Samples are grown by gas source molecular beam epitaxy (Riber 32P) using cracked phosphine and standard effusion cells for elements III, on undoped (001) GaP substrates. The strained layer growth is preceded by the growth of a 2000 Å thick GaP buffer layer at 600°C, exhibiting a (2×4) surface reconstruction. The substrate temperature is then lowered down to 520°C or 400°C during a 3 min growth interruption and the phosphine flux is vented when the substrate temperature reaches 550°C in order to keep the (2×4) reconstruction and to avoid any excess phosphorus accumulation on the surface. Once the substrate has reached the desired growth temperature, phosphine flux is turned on 3 s before the element III fluxes. Gallium and indium fluxes, determining growth rates and alloy compositions, are measured by RHEED intensity oscillations on GaAs and InP, before the growth. The growth rate of the strained GaInP layers lies in the 0.5–1 monolayer per second (ML/s) range except for the growth of InP for which it has been reduced to 0.1 ML/s. The PH_3 flow rate is 3 sccm for all compositions. 20 keV RHEED patterns are recorded along both the $[110]$ and the $[1\bar{1}0]$ directions. From the analysis of the RHEED patterns, we deduce the thickness H_{3D} for which the transition from a two-dimensional to a three-dimensional growth mode occurs from the intensity increase of a 3D Bragg diffraction spot. Ex situ AFM images are acquired with a Digital Nanoscope III system working in the tapping mode.

3. Results and discussion

During the growth of the strained layers at 520°C, whatever the In content, starting with the (2×4) GaP reconstruction, the RHEED pattern

changes to a (2×2) one as long as 2D growth occurs. We never observe this reconstruction during the growth of InP on InP or GaP on GaP or GaInP on GaAs in the same conditions. This reconstruction is then characteristic of the growth of GaInP on GaP. When the thickness increases, the (2×2) reconstruction progressively vanishes and the transition from a 2D to a 3D growth mode occurs. Fig. 1 displays the H_{3D} versus In content at 520°C. This evolution is rather similar to that observed in the GaInAs/GaAs system for $x \geq 0.3$ [7]. For low In content, differences appear between the two systems since around 520°C, 3D growth is not observed in the GaInAs/GaAs system for $x \leq 0.25$ [7,8] whereas it still occurs in the GaInP/GaP system for $x=0.2$. However, for this In content, we observe a great sensitivity of the relaxation mode to the growth temperature since for a substrate temperature lowered to 500°C, we do not observe anymore a 2D–3D growth mode transition and strain is relaxed via the formation of misfit dislocations. This indicates that in the temperature range considered, an In content of 0.2 represents the limit between the two relaxation modes. We now turn to a more detailed discussion of the film morphology after the onset of 3D growth and the specific case of the growth of InP on GaP.

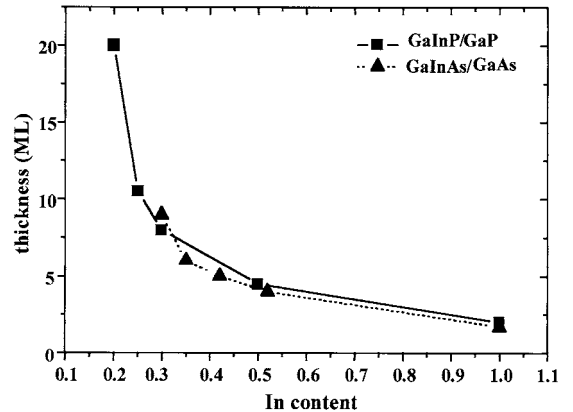


Fig. 1. Evolution of the H_{3D} thickness versus the In content x for GaInP/GaP at 520°C compared with GaInAs/GaAs (after Ref. [7]).

3.1. Surface morphology after the onset of 3D growth

At the 2D–3D transition and for $x \leq 0.5$, RHEED 3D Bragg spots are observed along the $[1\bar{1}0]$ direction whereas, along the $[110]$ direction, the RHEED pattern is very diffuse with arrow-head shapes. This indicates the formation of platelets elongated along the $[110]$ direction. This is confirmed by the AFM observations, evidencing the formation of wire-like structures along the $[110]$ direction (Fig. 2). This is markedly different from the InGaAs/GaAs case for which rather circular islands are observed for $x = 0.3\text{--}0.5$ [9].

Wire-like structures have been reported for the growth of InAs on InGaAs [10] but these structures are elongated along the $[1\bar{1}0]$ direction. In this case, the orientation of the wire-like structures is attributed to anisotropic surface diffusion of the impinging cations, typical of the As-stabilized arsenide surfaces [11]. More precisely, assuming the $(2 \times 4)\beta_2$ arsenide surface reconstruction [12], surface diffusion would be enhanced in the direction of the dimer rows, i.e. along the $[1\bar{1}0]$ direction, leading to the development of elongated islands along this direction.

Turning now to the phosphide growth, recent work [13] has shown that the (2×4) surfaces of InP and GaP are very different from that of GaAs. For GaP, two (2×4) reconstructions exist: under

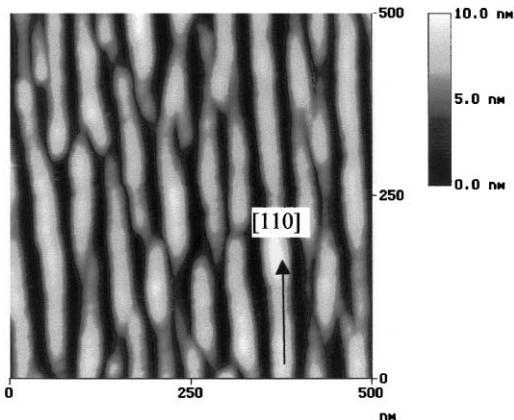


Fig. 2. AFM image of a 27 ML thick $\text{Ga}_{0.75}\text{In}_{0.25}\text{P}$ layer grown on GaP at 520°C .

UHV conditions, below 730°C , a P-rich one which looks like the $(2 \times 4)\beta_2$ GaAs one and a Ga-rich one above 730°C described by a mixed-dimer model. On the contrary, for InP, the (2×4) reconstruction appears to be solely an In-rich one with the same mixed-dimer model. Here, in the case of GaInP alloys, taking into account In segregation, the surface will be In enriched. Hence, we can infer that the (2×2) reconstruction we observe before the onset of 3D growth is more or less related to the InP (2×4) reconstruction. Apart from the mixed-dimer, this reconstruction is characterized by In-dimers along the $[110]$ direction. This would suggest a preferred surface diffusion along the $[110]$ direction which, in turn, would lead to the observed elongated islands along this direction.

3.2. InP on GaP

At 520°C , the RHEED pattern exhibits a (2×2) reconstruction before Bragg spots appear along both $[110]$ and $[1\bar{1}0]$ directions. The 2D–3D transition occurs abruptly at 2.1 MLs. This value is higher than that usually reported for the onset of 3D growth mode in the InAs/GaAs system, i.e. 1.7 MLs. The morphology of the film is very different too. As shown in Fig. 3a, after deposition of 3.3 MLs, two family of islands are clearly present. The first one corresponds to huge islands ($\sim 1\ \mu\text{m}$ wide, $\sim 2000\ \text{\AA}$ high) with a low density around $10^6/\text{cm}^2$. The second one is related to smaller dots ($\sim 1000\ \text{\AA}$ wide, $\sim 400\ \text{\AA}$ high) with a higher density around $10^7/\text{cm}^2$. These island sizes are very different from that observed in the InAs/GaAs system [14] where typical island are $200\text{--}300\ \text{\AA}$ wide and a few tens a \AA high with a much higher density above $10^{10}/\text{cm}^2$.

At 400°C , the RHEED pattern exhibits now a faint (3×2) reconstruction before the 2D–3D transition which is less abrupt than at 520°C but occurs around 1.7 MLs. As shown in Fig. 3b, the resulting island size and density are very different from the 520°C case too. The island size is more homogeneous with a mean width of $500\ \text{\AA}$, mean height of $150\ \text{\AA}$ and a density around $2 \times 10^{10}/\text{cm}^2$. From the island size and density, this 400°C

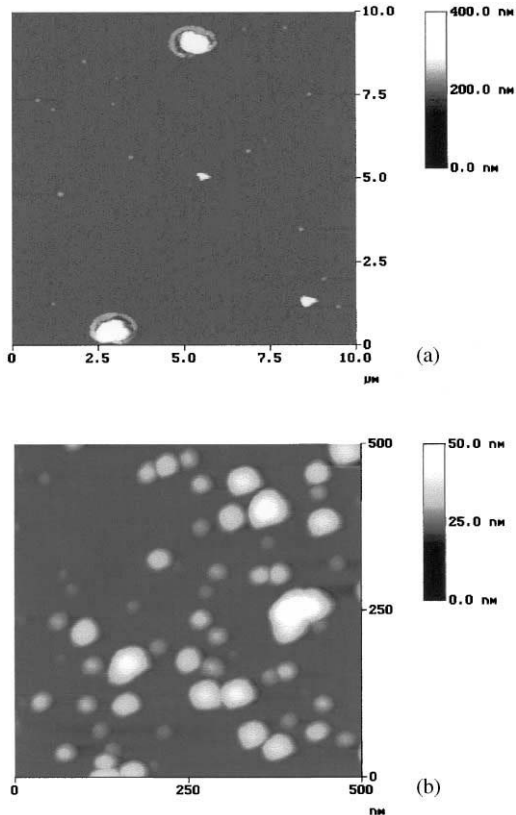


Fig. 3. AFM image after deposition of 3.3 MLs InP on GaP at 520°C (a) and 400°C (b).

growth looks closer to the InAs/GaAs case than the 520°C growth.

Since RHEED indicates that the surface reconstruction before 3D growth is different at 400°C and 520°C, the observed differences between 400°C and 520°C could be attributed to surface energy rather than to the surface mobility of the impinging species. Indeed, the fact that 3D islands are formed later at 520°C than at 400°C suggests that the (2×2) reconstruction observed at 520°C is more energetic than the (3×2) observed at 400°C. Then, the surface tension delays the 2D–3D transition at 520°C with respect to 400°C. In the same way, once the transition has occurred, the species mobility is greater at 520°C than at 400°C due to both the higher substrate temperature and to the surface tension. This is in good agreement

with the experimental behavior which evidences a more abrupt transition at 520°C.

4. Conclusion

In this work, we show that the evolution of the critical thickness for 3D growth versus In content in the GaInP/GaP system follows the same general trend than in the GaInAs/GaAs one. However, for In content below 0.5, 3D growth develops wire-like structures along the $[110]$ direction, never observed in the GaInAs/GaAs or GaInAs/InP systems. We show that this can be related to the (2×4) cation-rich surface reconstruction of phosphides. Finally, at 520°C, the growth of InP on GaP leads to the formation of large islands with a low density. At 400°C, the behavior looks closer like that observed in the InAs/GaAs system. We tentatively explain these results in terms of surface energy.

Acknowledgements

Authors are grateful to C. Coinon for his technical assistance and to C. Priester for fruitful discussions.

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