acta physica slovaca vol. 44 No. 4/5, 227 – 236 August 1994

KINETIC ROUGHENING DURING EPITAXIAL GROWTH OF IRON AND SILICON[1]

J. Chevrier[2], A. Cruz, I. Berbezier, J. Derrien
Centre de Recherche sur les Mecanismes de la Croissance Cristalline[3] CNRS Campus de Luminy case 913 13288 Marseille cedex 09 France

Received 13 April 1994, accepted 29 April 1994

In a molecular beam epitaxy chamber (MBE), we have performed epitaxial growths of iron on silicon (111) surface and homoepitaxial growths of silicon on silicon (111) surface. The thicknesses of the grown films were typically 500Å- 1000Å. We have observed a continuous and irreversible change of the Reflection High Energy Electron Diffraction (RHEED) patterns during the growth of iron at room temperature and of silicon at temperature below 400°C. This effect can be satisfactory explained by an increasing roughness of the surface during the growth. Annealing after growth leads in both cases to an irreversible flattening of the rough surface. These are experimental evidences for the occurrence of a kinetic roughness during epitaxial growth. In this article, a quantitative analysis of these RHEED pattern changes during epitaxial growth of silicon and of iron is presented. In both cases, the surface width s deduced from the RHEED patterns closely follows a power law $\sigma \propto h^{\beta}$. However the most reasonable values for β are around 0.25 for the growth of iron on silicon and $\beta \approx 1$ for the homoepitaxial growth of silicon (111). These results indicate that marked differences are found in the occurrence of kinetic roughening for these two different systems: iron on Si(111) or Si/Si(111).

1. Introduction

A wide variety of applications in fields like optics or microelectronics relies on the preparation and the control of thin films with thicknesses of about 100Å- 1000Å. In the past few years, the development of the MBE growth chambers for the preparation of thin films by epitaxial growth from the vapour phase [1] and the characterisation of the surface at an atomic level by high resolution direct space techniques like Scanning

[1] Invited lecture at MECO (Middle European CoOperation) 19, Smolenice, Slovakia, April 11-15, 1994

[2] Present address: Institut für Grenzflächenforschung und Vakuumphysik, Forschungszentrum Jülich, D 52425 Jülich, Germany

[3] The CRMC2 is associated to the Université Aix Marseille II

0323-0465/94 © Institute of Physics, SAS, Bratislava, Slovakia

Tunnelling Microscopy (STM) [2] have largely improved both the quality of the grown films and our understanding of the microscopic mechanisms involved in the epitaxial growth [3,4]. Subtle mechanisms of surface diffusion have been shown to be of a large influence on the appearing surface morphology during the first stages of the epitaxial growth [5].

In the limit of thicker films, the roughness of the growth front has to be investigated for both practical and fundamental reasons. For example, the quality of superlattices relies at least for part of it on the sharpness of interfaces. Clearly an intrinsic roughness appearing during the growth would then induce an alteration of the interface quality. On the other hand, theoretical analysis [6,7] have described how random deposition of atoms on surfaces triggers the development of a kinetic roughening during epitaxial growth. One of the motivation for the theoretical studies of the surface morphologies during epitaxial growth of thick films is the apparent lack of characteristic time and length in the growth dynamics. This is thought to lead the surface roughness to exhibit very general scaling relationships with time and space. A more complete theoretical description of the kinetic roughening during growth may be found in other contributions to this conference or in reference [6,7]. As the film grows, one expects the surface width s to be described by:

$$\sigma \propto h^{\beta} \quad (1)$$

where h is the film thickness. Depending on the underlying hypothesis on the adsorption and the desorption of atoms or on the characteristics of the surface diffusion, various theoretical descriptions have been developed. They lead to different description of the surface morphology and to different values of the exponent β. Both empirical and theoretical analysis led to the conclusion that a detailed experimental investigation of the surface morphology during the growth of thick films by vapour deposition is then definitely required. Although a precise and reliable characterisation of slow processes on long time scale and with space correlations over increasing distances is not an easy experimental task, several experimental analysis of this problem have been produced in the past few years [8-13]. The existence of a surface roughness specifically induced by the growth has been clearly observed. However it is noticeable that the values of the exponent β approximately lie between 0.2 and 1. One may also add that even a "supra linear" behaviour has been recently reported [14].

In view of the different parameters involved in the MBE growth, this variety of experimental results may not appear too surprising. Presumably a universal and over-simplified description of the surface response to the random deposition of atoms cannot reliably describe all the experimental findings. Indeed one has to take into account the detailed mechanisms of the surface diffusion and also the nucleation rate of atomic islands and their growth kinetics. As recently experimentally shown [4,5], different growth conditions can lead to very different shapes of the atomic islands nucleated during the growth on the surface. This is of importance as it has also been shown that the mass transport between terraces of different heights can depend upon the shape of the terrace edge [4,5]. The complexity of these surface processes suggests that depending on the exact experimental conditions, a variety of experimental results in the characterisation of growth front of thick films should be expected [15,16]. An experimental example of this wealth of behaviour seems to be found in the recent observation [13] of the variation of b with the temperature during the homoepitaxial growth of Cu on Cu(100). Finally silicon growth provides an example of the spectacular effects which can appear as further motivations for the study of the surface morphology of thick growing films. Indeed it is well known that a spontaneous amorphisation of silicon takes place when grown at low temperature (around room temperature) whereas at higher temperature (for Si(111), a reasonnable high temperature is 600°C - 700°C), a perfect growth with a flat surface can be performed for thicknesses up to several microns. Although this has been widely reported [1], a detailed analysis of this behaviour remains to be established.

2. Experimental

Silicon and iron growths on (111) oriented silicon wafers (two inch diameter) were performed in an MBE machine equipped with a silicon and an iron evaporation cell and a 10 kV electron gun for RHEED experiments. The base pressure of the MBE chamber at room temperature was between 1×10^{-10} and 2×10^{-10} torr. Cleaning of the initial silicon surface was based on two stages: i) an ex situ chemical etching based on the classical Shiraki procedure, ii) a desorption of the native oxide by heating the wafer at 800°C associated with an atomic beam of silicon directed onto the surface. Such a treatment ended up in a reproducible, clear 7×7 reconstructed silicon surface.

Our electron diffractometer is a conventional instrument comparable to many systems used to characterise epitaxial growth. The measurements presented here have been made possible owing to a systematic video acquisition of diffraction patterns. The camera we have used to record the RHEED patterns on the phosphorescent screen has a sensibility of 0.3 lux. Digitalisation of the video signal is done through an 8-bit computer card (Optiscan from Neotech), which means that the dynamic of this detector is limited to 256 grey levels. During the measurements, great care has been taken to avoid saturation either on the screen or during picture acquisition. The grazing angle of the electron beam with the surface was about one degree. The camera periodically took pictures of the RHEED patterns (the acquisition period could be changed from 1 picture/sec up to 1 picture/day) which were stored in the computer. Image analysis was then performed. Both intensities of diffraction spots and their widths (perpendicular and parallel to the sample surface) are numerically extracted. The complete digitalisation of diffraction patterns enabled us to analyse the position and the shape of the Bragg peaks.

During silicon growth, measurements of RHEED oscillations have been performed by a selective record of the reflected specular beam. The periodic changes of intensity of this small area in the RHEED screen could clearly be seen with bare eyes. One may notice that such a large variation was not observed in another part of the screen. Compared to the average reflectivity during growth, the variation of intensity during oscillations could be as large as 25% without any correction for the background. Figure 1 reports the RHEED oscillations measured during the silicon growth on a (111) silicon surface with a substrate temperature of $T \sim 350$°C. Analysis of 10 periods leads to a silicon flux of about 1 bilayer per minute (more precisely an increase of thickness $a_0/\sqrt{3} = 3.13$Åevery 65 seconds). Observation of large amplitude RHEED oscillations

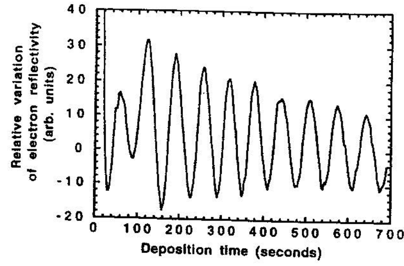

Fig. 1. RHEED oscillations observed by recording intensity of the specular beam during silicon growth at $T = 350°C$ on (111) silicon surface

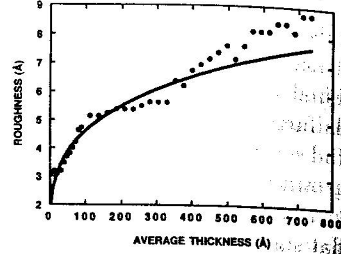

Fig. 2. Roughness perpendicular to the surface deduced from $\Delta G_\perp$ (FWHM of diffraction Bragg peaks measured perpendicular to the surface) during the iron growth. The solid line is the calculated surface width with $\beta = 0.25$ (see text)

during growth is a good indication that the cleaning procedure, the sample handling and the growth conditions are reasonable.

Beside RHEED oscillations, a second important effect is observed with bare eyes during silicon growth. Starting with the typical RHEED pattern of a flat ordered silicon surface, the condensation of diffraction streaks into conventional Bragg spots of silicon occurs. The same behaviour is observed during the growth of a thick iron film on silicon. In that case, the positions of Bragg peaks can be analysed in a way very much comparable to what is done for diffraction pattern produced by Transmission Electron Microscopy. This continuous change from a typical two dimensional diffraction pattern to a three dimensional pattern is a clear evidence for the occurrence of surface roughness. Indeed at the very beginning of the growth, the streaks observed in the diffraction pattern are the well known evidence that electron diffraction at grazing incidence is a surface technique. In the reciprocal space, the length of Bragg peaks perpendicular to the surface is primarily determined by the short penetration depth of the electron beam. If the surface becomes rough with typical roughness scales smaller than the inelastic mean free path of electron, then the surface width characteristic of this roughness is the effective sample size experienced by the electron beam. In the direction perpendicular to the surface, this imposes a length to the Bragg peaks. For the most important part, this is what determines the experimentally observed condensation of Bragg peaks during growth. The general fact that the width of the diffracted peak is inversely proportional to the size of the diffracting object provide a direct relationship between the perpendicular Full Width at Half Maximum of Bragg peaks, $\Delta G_\perp$, and the surface width, σ:

$$\Delta G_\perp = 2\pi C/\sigma \qquad (2)$$

C is a calibration constant that is determined using the distance between the 1×1 silicon streaks. In our particular case, the quantitative use of this relationship relies on two important approximations. A first important approximation is that the intensity along the diffraction streak is primarily determined by the surface structure and not by an instrumental effect. In the case of a clean and well ordered surface before growth, this is certainly far from to be correct. This has been shown in details [17]. For a very flat and ordered surface, the length of diffraction streaks is essentially determined by the intersection of the Ewald sphere and the weakly modulated RHEED streaks in the reciprocal space. In the case of very flat and ordered surfaces observed with a narrow beam of very small divergence, the streaks are very close to circular spots. For the results presented here, this has two main consequences: i) it is quite difficult to quantitatively estimate the initial roughness on the surface; ii) one has to make sure that during the growth, the apparent shape of RHEED streaks is dominated by the sample itself and not by the intersection with the Ewald sphere.

A second important approximation is that no other effect influences the perpendicular length of the diffraction peaks and that the measure of $\Delta G_\perp$ is directly related to the surface width. In the case of a perfect but rough crystal, this would be quite clear. In our RHEED patterns, a characterisation of the crystalline quality lies in the width of Bragg peaks parallel to the surface as there is no relaxation of the Bragg condition in this direction due to the limited penetration of electron. The lateral broadening of the Bragg peaks observed certainly means an equivalent broadening in all directions of the Bragg peaks. As the strong reduction of Bragg peak length, $\Delta G_\perp$, cannot be due to the crystal disorder, it clearly remains that the decrease of $\Delta G_\perp$ is strongly associated with the occurrence of surface roughness. Quantitatively it may appear difficult to separate the effect of roughness which tends to shorten the Bragg peaks and the appearance of a poor crystalline quality which gives rise to a broadening of Bragg peaks. Indeed in the final stage of the growth, the Bragg peaks are nearly circular. In that case, it would be difficult to safely separate the contribution of surface roughness and of atomic disorder from the peak width. This is only when one contribution is dominant that it is possible to quantitatively analyse the streak shape. The criterion we have used is that the streak length perpendicular to the surface must be much longer than the peak width parallel to the surface which is primarily determined by the crystalline quality. Therefore this is only between the two extreme regimes described here that one can use $\Delta G_\perp$ as a quantitative signature of the surface width.

3. Results.

A Growth of Fe on Si(111) at room temperature:

The deposition of iron on a Si(111) surface at room temperature in Ultra High Vacuum leads to the growth of an oriented but relaxed iron film. The characteristics of this epitaxial growth are described in reference [11]. In figure 2 is reported the variation of the surface width measured by RHEED following equation (2) during growth of iron on a Si(111) surface. This curve quantitatively shows the continuous effect of Bragg peak condensation observed in the RHEED pattern. Beside this basic result, one notices that a roughness in the nanometer range is found after the growth of an iron film of about 800Å. Although this roughness appears rather limited, the change of the RHEED during the growth is a strong effect and this result is reproducible. As mentioned in

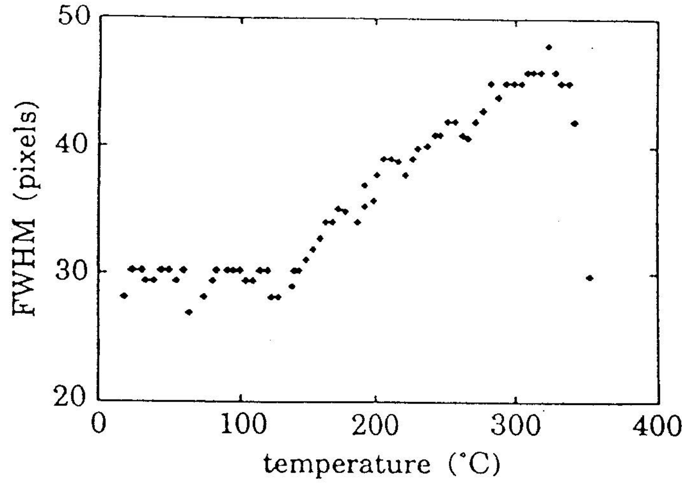

Fig. 3. Variation of $\Delta G_\perp$ during annealing after iron growth. The temperature increase is 1°C/min.

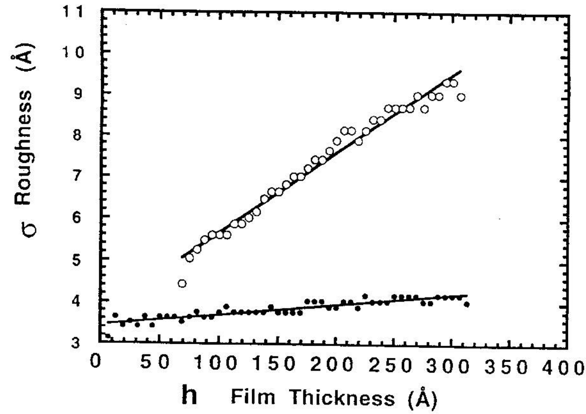

Fig. 4. Roughness perpendicular to the surface deduced from $\Delta G_\perp$ during the silicon deposition at $T = 300$°C (o) and at $T = 400$°C (full circles)

the previous section, it is unlikely that one can extract some information about the in plane structure of the roughness. It may only be possible to say that the inelastic mean free path known to be in the range of 100Åat the electron energy of 10keV, indicates an upper limit of the typical lateral scale of the observed surface roughness. The second point is the shape of the measured curved. The solid line follows the equation (1) with $\beta = 0.25$. For different measurements, the description of the experimental results using equation (1) is satisfactory and the adjusted exponent β is between 0.22 and 0.3.

In order to further characterise the roughened surface, we have followed the evolution

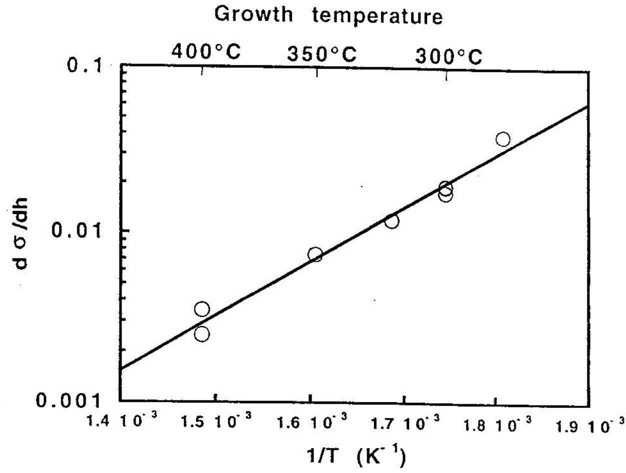

Fig. 5. $d\sigma/dh$ versus $1/T$. The straigth line is given by $\sigma = A\exp(E_f/kT)h$ where $A \sim 4.6 \times 10^{-8}$ and $E_f \sim 0.65$eV.

of the RHEED pattern during a controlled annealing. This is what appears in figure 3. The most important feature is an irreversible increase of the length of Bragg peaks. This occurs at a temperature which is quite consistent with the thermal activation of the iron surface diffusion. This means that the surface just after the growth is in a non equilibrium state. As atoms on the surface are thermally allowed to move, one observes a flattening of the surface.

All together the two points presented in figures 2 and 3 provide an experimental evidence for the occurrence of a rough surface in a non equilibrium state during the growth of iron on silicon. Also as shown in reference [11], we have further investigated how general is this behaviour. After a first annealing, it is possible to make subsequent cycles of iron growth and annealing. A clear feature is the reproducible condensation of Bragg spots during deposition and the increase of their length during each annealing.

B Growth of Si on Si(111) at different temperatures:

The variation of the surface width, $\sigma = 2\pi C/\Delta G_\perp$, versus the film thickness during homoepitaxy of silicon on Si(111) is presented in figure 4 for two different temperatures. For a total film thickness of about 500Å, this roughness is in the nanometer range. This is quite comparable to what has been observed for the iron epitaxial growth. Furthermore the importance of substrate temperature definitely appears in figure 4. At a growth temperature of $T = 400$°C, the occurrence of roughness is rather limited and this is even the limit we have been able to investigate by RHEED. At $T = 300$°C, the change of the surface is much faster and the kinetic roughening develops much more rapidly. Strictly looking at the experimental results in this range of film thicknesses, the best fit for the plot s versus h is obtained at all temperatures with a linear dependence

of the roughness with the film thickness. This linear behaviour is only approximate but this is a clear difference with the behaviour measured during the growth of iron .

Although the same kinetic roughening develops during epitaxial growth for two different systems like iron and silicon, clear differences are experimentally observed in details using the same RHEED technique and the same analysis of RHEED patterns. These differences in the development of kinetic roughening are also quite consistent with others studies [9,10,13] using different techniques. All these experimental results emphasize the fact that the occurrence of kinetic roughening during low temperature epitaxial growth is a general effect, but they also suggest that a unique and simple analysis for all surfaces does not seem to exist. A description of the microscopic mechanisms responsible for this difference is beyond the grasp of our experimental results. However they directly lead to the evidence that, depending of the systems, the surface diffusion and nucleation can relax the surface morphology (this corresponds to an apparent exponent β lower than 0.5) or amplify the surface roughness as shown by a measured exponent β larger than 0.5.

During epitaxial growth at different temperatures, the analysis of the surface morphology can provide further insights on the influence of surface mass transport in the development of kinetic roughening. The curves σ versus h measured at different temperatures present the same shape. This means that we can reliably extract the slope $d\sigma/dh$ at different growth temperatures. This is essentially what is reported in figure 5. The plot $\log(d\sigma/dh)$ versus $1/T$, the inverse of the growth temperature, strongly indicates that the general equation which describes the increasing roughness during growth is:

$$\sigma = A\exp(E_f/kT)h \quad (3)$$

with $E_f \sim 0.65$eV. Essentially this experimentally shows that, in the particular case of silicon, the appearance of the roughness during growth at low temperature is controlled by a thermally activated process on the surface. The thermally activated processes on surfaces one can think about during epitaxial growth, are basically the diffusion of silicon adatoms on a flat silicon surface and the nucleation of islands. The activation energy directly yielded by our analysis of roughness on the silicon (111) face is close to the measured activation energy $E_a \sim 0.67$eV reported in reference [3] for surface diffusion of adatoms on the (100) silicon face. Also recent measurements have reported an activation energy $E_a \sim 0.7$eV for surface diffusion of adatoms on the (111) silicon surface [18]. However, the identification of E_f with the activation energy E_a of adatom diffusion on silicon (111) surface cannot be reliably done at this point. Indeed an important experimental parameter in the control of the MBE growth process is the atomic flux on the surface. The influence of the silicon flux in the development of surface roughness during growth is not described by our results. This means that we do not know if whether or not Ef depends on the atomic flux. However a direct conclusion is that kinetic roughening is experimentally observed in a temperature regime where a large mobility of adatoms on a flat silicon surface clearly exists.

4. Conclusion

Although it is a general and characteristic behaviour of the surface morphology during epitaxial growth at low temperature, the specific and in situ investigations of kinetic roughening during growth of thick films (several hundred angstroms) are not numerous. It is however a central problem of molecular beam epitaxy seen as a dynamical process. For semiconductors, it can lead to specific and spectacular effects like progressive disordering or amorphisation which to our knowledge have not received a satisfactory explanation although they have been repeatedly observed.

In this paper by means of analysis of RHEED patterns, we have described the systematic occurrence of a roughness during the MBE growth of both iron on Si(111) and silicon on Si(111). Within the limits of the RHEED technique in the analysis of the surface roughness, we have determined some characteristics of this roughness. During the homoepitaxial growth of silicon on the Si(111) face, our experimental results indicate that the surface width vary linearly with the film thickness. This dependence of surface roughness with the film thickness is in marked difference with the results on the iron growth which have been obtained using exactly the same experimental method. This suggests that the details of mass transport mechanisms on the surface play an important role in the subsequent development of this roughness. Furthermore silicon homoepitaxy performed at different temperatures indicate that the developing roughness is controlled by a thermally activated mechanism. This activation energy is $E_a \sim 0.65$eV. The identification of the mechanism responsible for this activation energy cannot be inferred from these experimental results.

Finally for two different systems, we have shown that systematic acquisition and quantitative analysis of RHEED patterns reveal some major effects appearing during the epitaxial growth of thick films at low temperature. However many points have been left open due to the intrinsic limits of this technique. They certainly require further experimental investigations using different experimental methods. This is the case for the direct and in situ analysis of the crystal-amorphous transition during silicon growth, for the detailed description of roughness development in these two different regimes of growth and certainly also for the study of defect nucleation during silicon growth.

Acknowledgements: This work has been supported by the European Community through the Esprit Basic Research Action Project no. 3026. Grants from the French Ministère de la Recherche et de la Technologie and the PIRMAT-CNRS are also gratefully acknowledged (Contracts n. 88R0973 and 90S0249).

References

[1] Silicon Molecular Beam Epitaxy vol.1, 2 (1988) Edited by E. Kasper and J.C. Bean, (CRC Press, Inc. Boca Raton, FL)

[2] Max Lagally, Physics Today, November 1993 p 24

[3] Y.W. Mo, J. Kleiner, M.B. Webb and M.G. Lagally, Phys. Rev. Lett. 66, 1998, (1991)

[4] T.Michely, M. Hohage, M. Bott, and G. Comsa, Phys. Rev. Letters 70, 3943, (1993)

[5] R. Kunkel, B. Poelsema, L. K. Verheij and G. Comsa, Phys. Rev. Letters 65, 733, (1990); B. Poelsema, R. Kunkel, N. Nagel, A.F.Becker, G. Rosenfeld, L. K. Verheij and G. Comsa, Appl. Phys. A53, 369, (1991)

[6] R. Jullien, J.Kertesz, D.E. Wolf, eds. Surface Disordering: Growth, Roughening and Phase Transitions, Nova Science Publishers Commak N.Y. (1993)

[7] J. Villain, J. Physique I 1, 19, (1991)

[8] D. J. Eaglesham, H.-J. Gossmann and M. Cerullo Phys. Rev. Letters 65, 1227, (1990)

[9] Y.-L He, H.-N. Yang, T.-M. Lu and G.-C. Wang, Phys. Rev. Lett. 69, 3770, (1992)

[10] D. J. Eaglesham and G.J. Gilmer in reference [5]

[11] J. Chevrier, V. Le Thanh, R. Buys and J. Derrien Europhys. Letters 16, 737, (1991)

[12] J. Krim, I. Heyvaert, C. van Haesendonck and Y. Bruynserade, Phys. Rev. Letters 70, 57, (1993)

[13] H.-J. Ernst, F. Fabre, R. Folkerts, and J. Lapujoulade, Phys. Rev. Letters, 72, 112, (1994)

[14] D.P. Adams, S.M. Yasilove and D.J. Eaglesham, Appl. Phys.Letters 63, 3571, (1993)

[15] Zh Zhang, J. Detch and H. Metiu, Phys. Rev. B 48, 4972, (1993).

[16] I. Elkinani and J. Villain, submitted to Journal de Physique

[17] M.C. Tringides and M.G. Lagally in Reflection High Energy Electron Diffraction and Reflection Electron Imaging of Surfaces edited by P.K. Larsen and P.J. Dobson (Plenum New York 1988) p 523

[18] B. Voigtländer and A. Zinner Surf. Science Lett. 292, L775, (1993)