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Influence of kinetic roughening on the epitaxial growth of silicon

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Abstract. — The low temperature homoepitaxial growth of silicon has been probed *in situ* by Reflection High Energy Electron Diffraction (RHEED) in a Molecular Beam Epitaxy (MBE) chamber in the temperature range 250 °C-400 °C. The continuous change in the RHEED patterns during the growth of thick films (several hundred angstroms) shows the progressive appearance of a surface roughness during and after the decay of RHEED oscillations. This is a clear evidence for *kinetic roughening* in the case of silicon epitaxial growth at low temperatures on the Si(111) face. The surface width, σ , measured as the film thickness, h , is increased, can be described by : $\sigma(h, T) \sim Ah \exp(E_f/kT)$ with $E_f \sim 0.65$ eV. This is in marked difference with the kinetic roughening behavior measured during the growth of a metal like iron by means of the same RHEED technique (see Chevrier *et al.*, *Europhys. Lett.* **8** (1991) 737). Furthermore, an extrapolation of this behavior to epitaxial growths at higher temperatures ($T \sim 600$ °C-800 °C) suggests an effective influence of kinetic roughening in the determination of growth temperatures generally used for silicon MBE (i.e. the empirical usual growth temperatures $T_{epi} \sim 650$ °C). During growth at substrate temperatures between 250 °C and 400 °C, the nucleation of misoriented silicon islands takes place following the occurrence of kinetic roughening at the surface. In this range of temperatures, this suggests that the loss of epitaxy occurs on a rough surface through the proliferation of defects and of misoriented crystals.

1. Introduction.

In the field of silicon-MBE crystal growth, a large development of techniques has enabled one to grow high quality silicon crystals in ultra high vacuum conditions. Hence the growth of quasi-perfect homoepitaxial thin films is currently achieved in many laboratories. Nowadays, the number of studies produced in this field just prevent any attempt to collect a comprehensive bibliography. The major stakes and industrial applications are extensively describes in reference [1] Looking at the huge number of experimental results, one may think that the most

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important physical mechanisms involved in silicon-MBE are well known. Indeed the key experimental conditions to grow high quality silicon crystals are very well identified. However, there still exist major questions, with, to our knowledge, only limited answers.

For example, a crystal to amorphous transformation has been well identified during the MBE growth of silicon at low temperatures. This is a major feature of the silicon growth which prevents the growth of thick epitaxial films at low temperatures. However the mechanisms which spontaneously drive a single crystal of silicon into a completely disordered amorphous state during the growth at low temperatures, are still a matter of debate [2] and this is certainly an important issue in the understanding of epitaxial growth as a non-equilibrium process.

Furthermore, in order to establish the optimal experimental conditions for the MBE epitaxial growth of silicon, many attempts have been made to define a minimum temperature of the silicon crystal during silicon atom deposition [3]. The fact that MBE growth is a non-equilibrium process has only recently been fully included in the experimental analysis. This approach states that the continuous change of the crystal surface during the complete growth process must be taken into account to describe the final crystalline state [2, 4, 5]. A consequence of this point may be that the definition of the minimum temperature for the silicon epitaxial growth should be replaced by the determination of the thickest flat epitaxial film that one can grow at a given temperature. Such a statement is experimentally supported by the fact that at very low temperatures (i.e. around room temperature), the occurrence of the epitaxial growth has been clearly demonstrated [6]. The first stages of the silicon growth at low temperatures have definitely been shown to be of a reasonable epitaxial quality by transmission electron microscopy [2] before the epitaxial growth finally deteriorates as the thickness is increased. This shows that, at least in the low temperature regime, a continuous and irreversible transformation of the growing film structure is observed. The growth does not present a strick layer by layer growth mode and is not exactly periodic in times. This is only at higher temperatures (in the vicinity of the usual growth temperature), in good experimental conditions of growth, that no clear deterioration of the crystalline quality is observed throughout the whole film. In such a case, the electron reflectivity measured during the growths shows large RHEED oscillations with almost no noticeable damping even after deposition of several hundred silicon layers [7]. Incidentally, one may notice that if the observation of RHEED oscillations is a necessary step in the experimental control of MBE growth of metals or semiconductors, a detailed understanding of the surface characteristics controlling their amplitude and their damping only recently emerged [4]. Quite clearly real space techniques such as Scanning Tunneling Microscopy, Reflection Electron Microscopy and Low Energy Electron Microscopy have been at the heart of theses improvements. Together with Field Emission Microscopy [8], they have provided new results on the microscopic mechanisms of atomic transport on surfaces [4, 9, 6].

In this framework, the aim of this article is to experimentally describe the continuous changes of the RHEED patterns observed *in situ* during epitaxial growth of thick silicon films ($\sim 500 \text{ \AA}$) at different temperatures ($250^\circ\text{C} < T < 400^\circ\text{C}$). The interpretation of the diffraction peak shape will enable us to conclude that a kinetic roughness appears during growth and produces a surface in a non-equilibrium state. Together with the analysis of RHEED patterns, the ability to vary the growth temperature offers the possibility to investigate the development of this kinetic roughening and the loss of epitaxial growth in different conditions of atomic transport on the surface. This will also provide us with a quantitative surface description of how the epitaxial growth improves as the growth temperature is raised. Our complete results give some further insight into the reason why the definition of an epitaxial temperature in the field of silicon MBE was quite natural but also somewhat obscure as illustrated by a dispersion of about 200°C in its definition for the (111) silicon surface [3].

Furthermore theoretical analyses have described how random deposition of atoms on surfaces triggers the development of this kinetic roughening during epitaxial growth [10-12]. An important 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 a characteristic parameter like the surface width to vary as a power law of time. In a previous experimental investigation using the same RHEED technique, we have experimentally observed the occurrence of a kinetic roughening at the surface during the epitaxial growth of iron at low temperatures [5]. In this case, following the same experimental procedure as described in the next section of this article, the analysis of the Bragg peak shape in the RHEED pattern led us to characterize the continuous development of the kinetic roughness as the film thickness was increased. Directly deduced from the length of RHEED peaks, the roughness perpendicular to the surface, σ , was found to be linked with the film thickness, h , by the following relationship :

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

with $\beta \sim 0.25$. This power law is consistent with the expected scale invariance of the surface morphology during the development of kinetic roughening. Furthermore the exponent $\beta = 0.25$ is close to the value which is theoretically predicted for a kinetic roughening driven by the random deposition of atoms and relaxed by surface diffusion mechanisms. These results have been experimentally confirmed and completed by a recent study of iron epitaxial growth by means of *in situ* High Resolution Low Energy Electron Diffraction [13]. A behavior comparable to the one described by equation (1) has been found with an exponent $\beta \sim 0.22 \pm 0.022$. This is indeed quite consistent with our results although both studies have not considered the same iron crystalline face.

The experimental analysis of surface roughness occurring during the homoepitaxial growth of silicon on the silicon (100) silicon at low temperatures has revealed the occurrence of a similar kinetic roughening [14]. Analysis of this roughness has shown that the surface width can be reasonably described using equation (1). However the most consistent exponent with experimental results is $\beta \sim 1$. As pointed out in many references, it is reasonable to think that β should always be smaller than 0.5 if the surface diffusion relaxes the occurring roughness [10]. Therefore such an experimental finding basically implies that the roughness introduced by random deposition of atoms could be amplified by the mass transport mechanisms on the surface. This could find its origin in the experimental results on surface diffusion mechanisms deduced from Field Emission Microscopy [8]. A key point of these findings is the existence of a potential barrier at the upper part of the terrace edge. Whenever this barrier is present, it can lead during atomic deposition to an effective lack of mass transport between terraces of different heights. During the first stages of epitaxial growth, the dramatic influence of such mechanism on the occurrence of terraces with various heights has been demonstrated [4]. A numerical simulation presented in reference [14] has tested the efficiency of this restricted diffusion in developing surface roughness during epitaxial growth of thick films. The results suggest that a lack of atomic exchange between terraces of different heights causes a surface roughness during epitaxial growth which is much more important than that found when no barrier is taken into account.

Therefore it is a motivation of this article to investigate, in the same experimental conditions, the surface morphology observed during growth for two very different systems such as iron and silicon. This should enable us to provide some new insights into the *kinetic roughening of thick growing films* [13, 14].

2. Experimental.

Silicon growth on (111) oriented silicon wafers (two inch diameter) was performed in an MBE machine equipped with a silicon 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. The silicon source consisted of an evaporation cell using a thermally heated graphite crucible. The temperature control was better than 1 °C for both the substrate and the evaporation cell as it was measured on their own thermocouple. The accuracy of the temperature measurement was controlled on the crystallization of amorphous silicon and on the $7 \times 7 \leftrightarrow 1 \times 1$ transition at the silicon surface : the temperature shift should not be larger than about 20 °C. During silicon evaporation, the cell temperature was kept at 1360 °C. 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. For this particular study of silicon growth at low temperatures several growths could be achieved on the same sample with a good reproducibility of RHEED results provided that a long annealing at 800 °C was performed followed by the growth of a buffer of roughly 50 monolayers at about 800 °C. After this treatment, the RHEED pattern was very close to the initial one. However such a procedure could not be used when the signature in the RHEED pattern of nucleation of new and misoriented crystals did appear. Even after a long annealing at high temperatures, it is hardly possible to recover the usual RHEED pattern of silicon in the case. We shall also see that repeated cycles of growth and annealing ended up in an increase of the residual roughness. The influence on the developing roughness during the growth of these changing initial conditions will be analysed in the next section.

Our electron diffractometer is a conventional instrument comparable to many systems used to characterize 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 performed 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/s 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 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 were clearly visible to the naked eye. 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. In this article, we are not going to experimentally provide new insights into the growth mechanisms which determine the oscillations of electron reflectivity. It seems to us that it would be quite difficult to obtain new significant results on the sole basis of electron diffraction at grazing incidence. An experimental basis of this remark is the observation that the damping of RHEED oscillations measured during growth varies from sample to sample with different initial surface treatments although the initial RHEED patterns

are hardly different. Also in the case of our experimental set up, i.e. a non-rotating sample with a diameter of two inches, even a small non-homogeneity of the silicon beam would induce an apparent damping of the RHEED oscillations. In our case, this last point almost completely prevents any attempt to analyse the damping of RHEED oscillations quantitatively. Motivation for the measurement of these oscillations is twofold. First, this enabled us to determine the silicon flux. Figure 1 reports the RHEED oscillations measured during the silicon growth on a (111) silicon surface with a substrate temperature of $T \sim 350^\circ\text{C}$. Analysis of 10 periods leads to a silicon flux of about 1 bilayer per minute (more precisely an increase of thickness $a_0/\sqrt{3} = 3.13 \text{ \AA}$ every 65 s). Also the observation of large amplitude RHEED oscillations during growth is a good indication that the cleaning procedure, the sample handling and the growth conditions are reasonable.

Fig. 1. — RHEED oscillations observed by recording intensity of the specular beam during silicon growth at $T = 350^\circ\text{C}$ on (111) silicon surface.

Beside RHEED oscillations, a second important effect is visible to the naked eye during silicon growth. Starting with the typical RHEED pattern of a flat ordered silicon surface, the condensation of RHEED streaks into conventional Bragg spots of bulk silicon occurs. In this case, the positions of Bragg peaks can be analysed in a way very much comparable to what is done for diffraction patterns 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 in the reciprocal space, the length of reciprocal rods perpendicular to a flat initial surface is primarily determined by the short penetration depth of the electron beam. As the surface becomes rough, the electron beam goes through surface bumps and the reciprocal rods condense towards bulk diffraction peaks (An illustrated description of these mechanisms can be found in ref. [15]). In the direction of the reciprocal space, perpendicular to the surface, the surface roughness imposes a limited length to the diffraction peaks. The general fact that the width of the diffracted peak is inversely proportional to the size of the diffracting object provides a direct relationship between the perpendicular Full Width at Half Maximum of the diffraction peaks, $\Delta G_\perp$, and the surface width, σ , which is characteristic of the surface roughness and corresponds to the effective sample size experienced by the electron beam :

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

where C is a calibration constant that is determined using the distance between the 1×1 silicon streaks. The use of this quantitative relationship relies on restrictive approximations. A description of the RHEED technique with its main limitations can be found in reference [15]. In the particular case of silicon growth at low temperatures, limitations due to two severe approximations are directly observed in the RHEED patterns. A first approximation is that the measured intensity along a RHEED streak is primarily determined by the surface morphology and not by an instrumental effect. This especially supposes that the intersection of the reciprocal space with the Ewald sphere has essentially no influence on the value of $\Delta G_{\perp}$. In the case of a clean and well ordered silicon surface before growth, this is certainly far from to be correct. This has been shown in detail in [15]. For a very flat and ordered surface, the lengths of RHEED streaks are essentially determined by the intersection of the Ewald sphere and the weakly modulated reciprocal rods. As seen in figure 2, this intersection is the origin of large circles formed by short RHEED streaks in silicon diffraction patterns (see also [16]). In the case of very flat and ordered surfaces observed with a narrow beam of very small divergence, this effect can be so large that one observes 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 silicon growth, the apparent shape of RHEED streaks is dominated by the sample itself and not by the intersection with the Ewald sphere. During deposition of the first few layers deposited at growth temperatures around $T \sim 300^{\circ}\text{C}$, the diffraction pattern is deeply changed. The RHEED streaks due to the 7×7 reconstruction are quickly weakened and those characteristic of the 1×1 silicon structure are laterally broadened. A consequence of this is a longer intersection of diffraction streaks with the Ewald sphere resulting in long streaks in the RHEED patterns. This is qualitatively the RHEED pattern expected from a flat surface with a deteriorated crystalline order. The length of these RHEED streaks at the beginning of the growth shows the maximum intersection between the modulated reciprocal rod space and the

Fig. 2. — RHEED pattern observed on a clean (111) silicon surface before growth with electron beam along $[1\bar{1}0]$ azimuth.

Ewald sphere. This indicates that the roughness measurement is reasonable as long as the analysis is confined to Bragg spots close to the center of the reciprocal space. The results presented here have all been acquired under those conditions. The Bragg spots generally used for our experiments are those with Miller indices 113 and 220 (the electron beam is along a $[1\bar{1}0]$ azimuth and the axis perpendicular to the surface is the $\langle 111 \rangle$). A further consequence of these experimental limitations is the range of growth temperatures that we can use for a study of kinetic roughening by means of RHEED. Indeed above a growth temperature of $T \sim 400^\circ\text{C}$, evidence for the appearance of roughness in the RHEED patterns during the epitaxial growth is limited. This prevents a quantitative analysis of the roughness based on grazing incidence electron diffraction.

A second approximation is that the variation of $\Delta G_\perp$ can be analysed as the quantitative signature of only one mechanism: the occurrence of the surface roughness. In the case of the growth of a perfect crystal, the variation of $\Delta G_\perp$ in the reciprocal space would be directly connected to the occurrence of a surface roughness. In our RHEED patterns, a characterization 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. In the temperature range we have investigated, a lateral broadening of the Bragg peaks is observed which shows that the crystalline quality deteriorates during the growth. This 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 a 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 this case, it would be extremely 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 analyse the streak shape quantitatively. 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 [17, 5]. Therefore the two extreme regimes described here impose constraints on the use of $\Delta G_\perp$ as a quantitative signature of the surface width.

3. Results.

3.1 GENERAL OBSERVATIONS. — In figure 3, pictures of the full RHEED screen are presented at different stages of the growth for a substrate temperature at $T = 280^\circ\text{C}$. The electron beam is along a $[1\bar{1}0]$ silicon azimuth. Signatures of the major effects which are quantitatively investigated in this article are clearly seen in this figure 3. Compared to figure 1, picture a of figure 3 taken after deposition of a few layers on the surface shows the disappearance of the 7×7 diffraction streaks with long and rather broad 1×1 streaks. Such a change of the diffraction pattern is consistent with a limited roughness and disorder within the surface. Further deposition (picture b of Fig. 3) leads to a complete disappearance of the 7×7 reconstruction and to a condensation of Bragg spots. Such an effect is seen for all spots of the diffraction pattern. This is a strong evidence for the continuous occurrence of a surface roughness during the growth. Qualitatively such a result is very much comparable to what is observed in other cases [5, 6]. In this range of temperatures further silicon growth clearly induces new spots in the diffraction pattern (picture c of Fig. 3). These new spots call for several remarks: 1) these are new, localised and well defined spots. No diffraction rings are observed at the same time in the diffraction pattern. 2) These new spots do not appear at the

Fig. 3. — Characteristic diffraction patterns observed during deposition of silicon at $T = 280^\circ\text{C}$.

very beginning of the growth but after development of some roughness on the surface. iii) This is a highly reproducible result in this range of temperatures. However, for growth temperatures above 400°C and for the thicknesses investigated, the occurrence of these new Bragg spots is hardly seen. This observation will be more quantitatively developed in a following paragraph with the determination of the crystallographic orientation of these new crystals on the substrate. Already, this is an evidence for the nucleation of new crystallites on the surface and for an original mechanism leading to the loss of epitaxial growth during silicon deposition on silicon (111) surface in the temperature range 250°C - 400°C . At lower temperatures such a behavior is not clearly observed: the whole original diffraction pattern disappears after condensation of streaks into Bragg spots. This means that the complete disappearance of the diffraction pattern follows the occurrence of a rough surface. After this irreversible transformation, only large and diffuse rings are left on the screen. This is the commonly reported evidence for the RHEED observation of amorphous silicon. An important experimental observation already reported at other growth temperatures [6, 7] is that, after the atomic flux is stopped, no change of the silicon surface is observed at the growth temperature (always

below 400 °C). However heating the sample at a higher temperature (around 700 °C) induces an irreversible flattening of the surface and the diffraction evidence for the 7×7 silicon reconstruction is recovered.

These major and general observations of the kinetic roughening during the silicon growth at low temperatures are consistent with what we have previously reported for the growth of iron. This emphasizes how much general this effect is. The specific and quite important effect observed for silicon is the loss of the epitaxial growth of a single crystal. This is in marked difference with the epitaxial growth of iron [5] for which no qualitative loss of the epitaxial growth was observed even though a comparable kinetic roughening was observed as seen by the RHEED technique.

3.2 QUANTITATIVE ANALYSIS OF KINETIC ROUGHENING. — The variation of the surface width, $\sigma = 2 \pi C / \Delta G_{\perp}$, versus the film thickness is displayed in figure 4 for two different temperatures. The first observation concerns the absolute value of the roughness measured. For a total film thickness of about 500 Å, this roughness is in the nanometer range. This is quite comparable to what has been observed for the iron epitaxial growth. It is also very close to what is reported for the growth of silicon on silicon (100) surface in the equivalent temperature range [14]. These measurements have been performed in direct space by Transmission Electron Microscopy whereas our results have been obtained by means of a reciprocal space technique. A second point is that 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 quicker 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 σ versus h is obtained at all temperatures with a linear dependence of the roughness with the film thickness. This is similar to the experimental result obtained during the growth on the (100) silicon surface [14]. This linear behavior is only approximate but this is a clear difference with the behavior measured during the growth of iron [5].

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 other studies [13, 14] using

Fig. 4. — Roughness perpendicular to the surface deduced from the FWHM of diffraction Bragg peaks during the silicon deposition at $T = 300$ °C (○) and at $T = 400$ °C (●).

different techniques. Therefore 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 on 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 into 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 $\text{Log}(d\sigma/dh)$ 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 $E_f \sim 0.65 \pm 0.07$ eV. Essentially this shows experimentally that, in the particular case of silicon, the appearance of the roughness during growth at low temperatures is controlled by a thermally activated process on the surface. The thermally activated processes on surfaces one can think of 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 [9] for surface diffusion of adatoms on the (100) silicon face. Recent measurements have also 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

Fig. 5. — $d\sigma/dh$ versus $1/T$. The straight 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.

development of surface roughness during growth is not described by our results. This means that we do not know whether or not E_f 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.

In order to link these experimental results with the common practice in MBE growth of silicon, we extrapolate equation (3) to a larger range of temperatures than the one we have measured. We take as a conventional and reasonable definition of a good quality silicon film grown by MBE, a roughness lower than 1 Å. Although such a low value is in fact not measurable by RHEED and would certainly correspond to a diffraction pattern hardly changed by the growth, this definition provides a reliable criterion. A slight change in the accepted final roughness of 1 Å would not significantly alter this analysis. Figure 6 shows the maximum thickness one can grow with that constraint of low roughness at different growth temperatures. This curve clearly indicates that growth temperatures of about 600 °C-700 °C are needed to obtain flat surfaces after growth of thick silicon films (about 5 000 Å-10 000 Å). It is clear that this plot is only an extrapolation of measurements performed at lower temperatures and that the film quality is not only determined by the surface flatness. Furthermore since we are dealing with kinetic effects induced by random deposition of atoms, one would expect these results to exhibit a dependence not only with temperature but also with the number of atoms coming on the surface. Using an evaporation cell, the silicon flux we have used is between one or two orders of magnitude lower than the usual fluxes used in MBE equipped with electron guns. However, within our experimental conditions, the conclusion is that it is necessary to grow silicon at temperatures higher than 600 °C to remove significant effects of kinetic roughening in the growth of thick (111) silicon films. Despite the aforementioned limits of our experimental investigation, it is remarkable that this is what is currently done in silicon MBE growth. For example, in reference [3], epitaxial temperatures for (111) silicon are reported to be around 520 °C and 700 °C depending on experimental characterizations used to define this epitaxial temperature. Also, as the film thickness in MBE growth required for applications must be in the range 1 000 Å-10 000 Å, the curve in figure 6 can easily be approximated by a

Fig. 6. — Estimate from figure 5 of the growth temperature range needed to grow an epitaxial silicon film with a thickness in the micron range and with a roughness lower than 1 Å. The inset with the experimental result shows the same curve for lower thicknesses and therefore lower growth temperatures.

constant growth temperature of 700 °C for any desired thickness of a flat epitaxial silicon film. This is represented by the dotted line in figure 6. This provides a plausible explanation for the determination of an epitaxial temperatures and suggests that the kinetic roughening occurring during silicon epitaxial growth is one of the parameters which implicitly control the determination of conventional experimental conditions of silicon epitaxial growth.

In figure 7, the development of roughness during epitaxial growth at $T = 300$ °C is presented for different initial surface morphologies. The main characteristic of these initial surfaces is that they have been obtained after several cycles of growth at low temperatures, annealing at high temperatures ($T \sim 800$ °C) and growth of a buffer at $T \sim 700$ °C. The experimental evidence is that the variation of σ with h does not depend on the initial roughness. Indeed the slope $d\sigma/dh$ is found to be the same for different residual roughnesses measured by RHEED. Although we are limited in our experimental characterisation of surfaces, it is quite remarkable that the original surface state issued from growth and thermal annealing is not relevant in the subsequent development of kinetic roughening. In figure 7, a flattening of the curve σ versus h is clearly observed for the largest values of the film thickness. In this range of thicknesses, the RHEED patterns show the large size of the secondary diffraction spots due to new crystallites and diffraction spots with lengths close to their widths. This apparent saturation of kinetic roughening is reproducible. Its interpretation on the strict basis of experimental results is rather complicated. As described in the previous section about experimental conditions, this flattening may only reveal a limit of the electron diffraction technique at grazing incidence in the analysis of rough surfaces.

Fig. 7. — Roughness perpendicular to the surface deduced from the FWHM of diffraction Bragg peaks during the silicon deposition at $T = 300$ °C. This characterizes the effect of cycles of growth and annealing on the residual roughness and on the appearing roughness during the growth (see text)

3.3 CREATION OF DEFECTS DURING EPITAXIAL GROWTH. — Figure 8 clearly shows that the diffraction pattern obtained after the growth of a thick silicon film in the temperature range 300 °C-400 °C, is very different from that given in figure 1. The spotty diagram provides clear evidence for the deterioration of the epitaxial growth. The new Bragg spots seen on the pattern are a clear signature of well defined defects in the film. When the new spots appear, the absence of continuous rings shows that randomly distributed silicon grains are not present. Indexation of the complete diffraction pattern along a $[1\bar{1}0]$ azimuth can be done at this first

Fig. 9. — Measurement of the intensity of the appearing Bragg spot *versus* the deposition time.

epitaxial growth on (111) silicon surface different from the usual crystal-amorphous transition observed at lower temperatures and which appears to be induced by the kinetic roughening. This crystallographic identification of the initial nucleation of misoriented crystals during growth on (111) silicon surface also provides some further information on the increasing defect density reported in reference [19] during epitaxial growth on silicon (111) in the same range of temperatures.

Conclusion.

Although it is a general and characteristic behaviour of the surface morphology during epitaxial growth at low temperatures, 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 experimentally observed the systematic occurrence of a roughness during the growth of silicon at different temperatures. 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 varies linearly with the film thickness. This dependence of surface roughness with the film thickness is in marked difference with our previous 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 growth experiments 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 is beyond these experimental results. We have made an extrapolation of the temperature variation of the surface width to relevant temperatures for high quality silicon growth. This suggests that the occurrence of a kinetic roughness may be a relevant parameter in the determination of the usual experimental conditions for the MBE growth of silicon.

After the occurrence of a significant amount of roughness, we have observed the spontaneous breakdown of the epitaxial growth. At very low temperatures, an amorphous phase readily follows the loss of the crystalline order. In medium range temperatures (250 °C-400 °C), we have identified a progressive appearance of specific crystalline defects. Whatever the detailed mechanism, this epitaxial breakdown is a consequence of crystal growth by means of a process far from equilibrium. To our knowledge, despite the significant remarks of reference [14], a satisfactory description of silicon disordering during silicon MBE growth is not established. It is then an aim of this paper to add some further experimental insights into the knowledge of this problem. We have found that as the temperature is raised, this effect seems to disappear progressively. RHEED studies of the surface morphology suggest that the occurrence of a kinetic roughness is a key point in the breakdown of epitaxial growth. 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 temperatures. 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.

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$$\Delta G_{\lambda} = \sqrt{\Delta G_{\perp}^2 - \Delta G_{\parallel}^2}.$$

In the linear part of the curve σ versus h , a usual experimental value for the ratio $\Delta G_{\perp}/\Delta G_{\parallel}$ is about 3 or larger. This means that the difference between $\Delta G_{\perp}$ and ΔG_r should be about 5 % or less. This gives an indication of how approximate our method is. As the ratio $\Delta G_{\perp}/\Delta G_{\parallel}$ becomes close to 2, with a difference between $\Delta G_{\perp}$ and ΔG_r becoming larger than 10 %, one experimentally observes an apparent saturation in the increase of the surface roughness. As mentioned in the text, this saturation may indeed be the sign that the crystalline coherence is, at this stage, the important factor to determine $\Delta G_{\perp}$ with a limited effect of the surface width.

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