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Surface morphology of low temperature grown GaAs on singular and vicinal substrates

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Abstract

The evolution of the surface morphology of epitaxial GaAs layers grown at low substrate temperatures (LT-GaAs) on singular and vicinal (001) GaAs substrates is studied by means of kinetic Monte-Carlo simulations. The simulation model includes the effects of Ehrlich–Schwoebel barriers at step-edges as well as anisotropic surface diffusion. We find that the surface morphology is dominated by a pattern of elongated growth mounds, which are organized into columns parallel to [$\bar{1}$10]. The formation of this pattern is gradually suppressed on vicinal substrates as the misorientation angle increases. Simulated surface morphologies are compared to atomic force microscopy measurements on LT-GaAs epilayers grown on singular GaAs(001) substrates at different temperatures and good quantitative agreement is found. We propose to use vicinal substrates for LT-GaAs growth in order to overcome the known problem of epitaxial breakdown above a certain epitaxial thickness. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: LT-GaAs; Kinetic Monte-Carlo; Vicinal

1. Introduction

The surface morphology of thin films and its evolution during epitaxial growth has been the subject of a large number of experimental and theoretical studies in recent years [1]. The physical phenomena that take place in a non-equilibrium process like epitaxy are of fundamental importance from the point of view of condensed matter physics. Furthermore, there is also great technological interest in the subject, with respect to the improvement of thin film surfaces and interfaces as well as to the use of growth related surface features in fabricating lateral nanostructures. Epitaxial growth at low substrate temperatures and the corresponding surface morphology is of particular interest in the case of semiconductors, due to the elimination of unwanted effects, like dopant segregation and interface interdiffusion. Additionally, growth of III–V semiconductors at low substrate temperatures leads to non-stoichiometric materials, which exhibit unique properties and have found many important applications [2].

A very interesting phenomenon, common to a broad range of epitaxial systems, is the formation of three-dimensional (3D) features called mounds during growth on an initially flat singular surface [3]. Mound formation has been attributed to the presence of the so-called Ehrlich–Schwoebel (ES) diffusion barriers [4,5], which inhibit the downward movement of adatoms at surface step-edges. Due to the ES barrier adatoms are hindered from stepping down from a 2D island, thus, there is an increased probability that new 2D islands will nucleate on top of already existing ones. The repetition of this process leads to the multilayer structures called growth mounds.

In a previous publication [7], we reported that the formation of growth mounds in homoepitaxial GaAs(001) is reentrant. In particular, mounds appear on the surface of MBE grown GaAs(001) at high ($T \sim 600$ °C) and low ($T \sim 200$ °C) substrate temperatures, while at intermediate temperatures the surface appears smooth. This behavior has been attributed to an enhancement of adatom mobility at low temperatures through the condensation of excess As on the surface, which functions as a surfactant modifying the surface kinetics of Ga [6,7]. In this work we extend our investigation of the low temperature mounding behavior of GaAs, by comparing the evolution of the growth mound pattern with respect to substrate temperature to the results of the detailed Monte-Carlo growth simula-

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tions. Furthermore, we discuss the relevance of the effect of mound formation to the breakdown of perfect epitaxial growth when a certain epitaxial layer thickness is exceeded, and we propose a possible way of overcoming the epitaxial thickness limit by growth on vicinal substrates.

2. Simulation model

The growth model invoked here is based on previous work about kinetic Monte-Carlo simulations [8,9] of homoepitaxy in metallic and semiconductor systems. The basic assumptions are: an fcc(001) substrate with neither bulk vacancies nor overhangs allowed; atoms are deposited randomly on the substrate at a rate f and are finally incorporated at fourfold hollow (4FH) sites; and only the group-III species kinetics are considered, since growth is limited by the supply of Ga under the given MBE conditions. This justifies also the use of an fcc lattice, since we consider only the Ga sublattice. The incorporation of a deposited atom proceeds in two steps:

1. *Downward funneling*. If the atom impinges on top of a surface atom or at a step-edge, it moves downward to one of the neighboring 4FH sites chosen randomly [10]. Otherwise, if the atom impinges directly on a 4FH site, no movement takes place at this stage. This process corresponds to the fact that initially deposited adatoms will dissipate some of their condensation energy. Šmilauer and Vvedensky [9] have extensively used a similar incorporation mechanism, and have shown that it is crucial for the simulation of GaAs growth. This implementation of funneling has been generally used in simulations of metallic systems, and we adopt it here because it is naturally formulated for the fcc lattice and does not require the introduction of free parameters. However, a microscopic interpretation is still lacking for semiconductor surfaces.
2. *Diffusion*. Once the atoms have reached a 4FH site they are allowed to diffuse on the surface. The atoms may hop to one of their neighboring 4FH sites along the next-nearest-neighbor direction. The hopping rate is given by $R(E,T) = R_0 \exp(-E/k_BT)$, where E is the diffusion barrier and the prefactor R_0 is assigned a value of $R_0 = 2k_BT/h$, where h is Planck's constant [9]. The diffusion barrier comprises a substrate contribution E_a, a contribution E_n from each in-plane bond and an additional barrier E_s when an adatom goes down a step, which corresponds to the ES barrier. In order to account for anisotropic surface morphologies, we introduced an anisotropy in the substrate contribution to the barrier. Thus, we distinguish two different substrate barriers along the diffusion directions on the (001) surface, $E_a^{[110]}$ and $E_a^{[\bar{1}10]}$ and we define an anisotropy factor $\alpha = \exp\{-(E_a^{[110]} - E_a^{[\bar{1}10]})/k_BT\}$.Simulations were performed on square substrates with periodic boundary conditions. We used the values $E_a^{[110]} = 0.7$ eV, $E_s = 0.1$ eV, $E_n = 0.23$ eV, and $\alpha = 1.33$ for the model parameters. The deposition rate was kept constant at 1 monolayer s^{-1}.

3. Results and discussion

3.1. Singular substrates

Fig. 1 shows atomic force microscopy (AFM) images of the surface of 1 μm thick GaAs epilayers grown by MBE at low substrate temperatures. The sample preparation and growth process is the same as described in a previous publication [6]. As can be seen in the figure, a growth pattern emerges as the growth temperature T is reduced. The pattern consists of mounds elongated along [$\bar{1}$10], which are organized into columns running roughly parallel to [$\bar{1}$10]. The height of the mounds increases with decreasing T, while their lateral dimensions decrease and they become more asymmetric.

Fig. 2 shows gray-scale images of surface morphologies obtained by simulating the growth of 1 μm thick GaAs epilayers at different growth temperatures. A

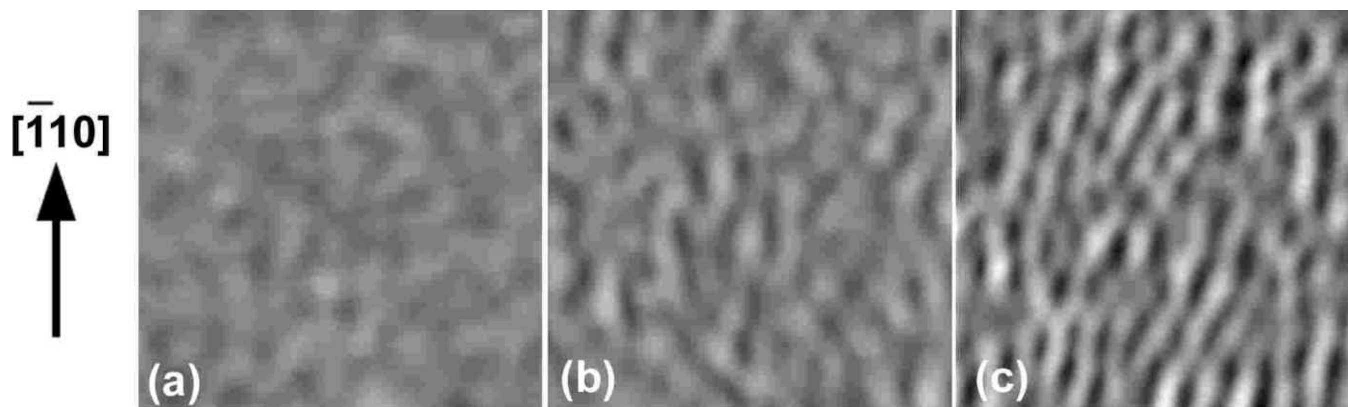

Fig. 1. AFM images of 1 μm thick GaAs epilayers grown at different substrate temperatures: (a) 260; (b) 230; and (c) 210 °C. The area of the images is 500 × 500 nm and the gray scale corresponds to a height difference of 8 nm.

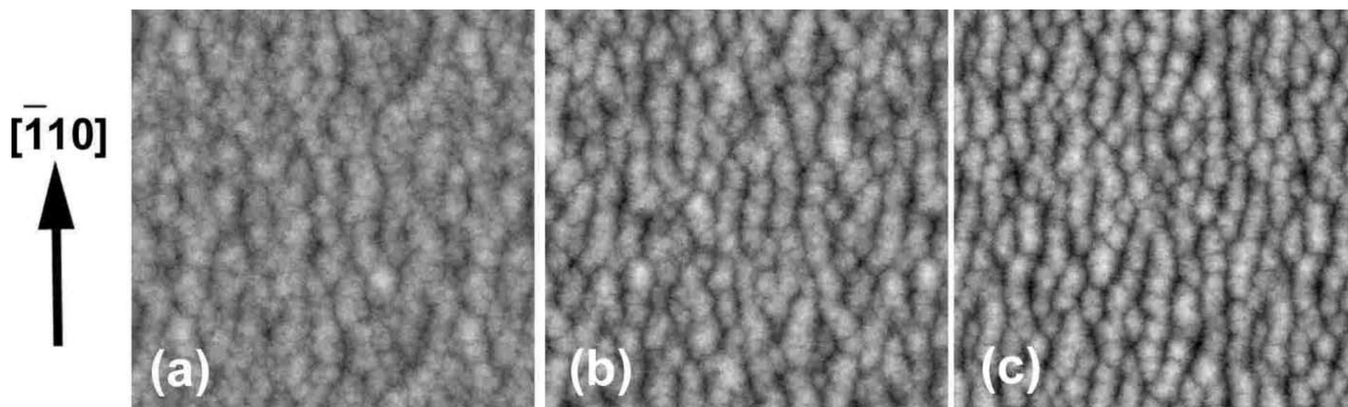

Fig. 2. Gray-scale images of the surface morphologies obtained from growth simulations of 1 μm thick GaAs(001) layers at different growth temperatures: (a) 260; (b) 230; and (c) 210 °C. The area of the images is 500 × 500 nm and the gray scale corresponds to a height difference of 8 nm.

direct comparison between the simulation results of Fig. 2 and the AFM images of Fig. 1 shows that our growth model reproduces the main features of the surface morphology, i.e. the appearance of a growth mound pattern, elongated along [$\bar{1}$10], which fades out as the temperature is increased.

For a quantitative analysis of the surface characteristics, we calculate the height–height correlation function $G(x, y) = (1/2)\langle\tilde{h}(x+x', y+y')\tilde{h}(x, y)\rangle_{(x', y')}$, where $\tilde{h}$ is the height relative to the average height, $\langle\cdots\rangle$ denotes an averaging over the entire image area, and x and y are parallel to [110] and [$\bar{1}$10], respectively. The root-mean-square (rms) width (roughness) is given by $w = \sqrt{G(0, 0)}$. The position of the first maximum of $G(x, 0)$ corresponds to the average distance between growth mounds parallel to [110], thus, twice the lateral mound radius $R_c^{[110]}$. The data extracted from the height–height correlation functions are presented in Fig. 3 as a function of growth temperature. A direct comparison between the experimental and simulation results shows that our growth model reproduces the main features of the surface morphology. The agreement for the radius $R_c^{[110]}$ is excellent, while for the surface width the behavior is described qualitatively and the simulation yields realistic values. Thus, we conclude that the ES step-edge barrier picture may describe satisfactorily the surface morphology of epitaxial GaAs on (001) singular substrates at low growth temperatures. It should be noted that the values of E_a and E_s used here are smaller than those for the high temperature case (1.54 and 0.175 eV, respectively [9]). This modification of adatom kinetic properties has been attributed to condensation of excess As on the surface at low temperatures [6,7], which acts as a self-surfactant. Ga adatoms are first deposited on top of this excess As layer, on which they have a lower diffusion barrier. Since such a surfactant effect is temperature dependent, one would expect that the diffusion barriers should also depend on temperature. In order to keep our model as simple as possible, we did not consider temperature dependent barriers. However, the very good agreement between simulation and experiment shows that this effect is of minor importance.

3.2. Vicinal substrates and epitaxial breakdown

As a consequence of mound formation the surface of LT-GaAs samples have an increased roughness. Eaglesham et al. [11] proposed that surface roughness might be the origin of the well-known problem of epitaxy breakdown in low-temperature growth, which occurs above a limiting epitaxial thickness. This breakdown begins with the nucleation of the so-called ‘pyramidal defects’ [12], which appear as bunches of stacking faults and twins on (111) planes. Since we observe the formation of a regular anisotropic pattern in the developed surface roughness, we assume that there is a relation between the roughness pattern and the pyramidal defects. Consequently, a way to overcome the epitaxial breakdown limitations would be to suppress the formation of the mound pattern.

In systems exhibiting mound formation this may be accomplished by the use of vicinal substrates. The development of mounds on a vicinal surface is governed

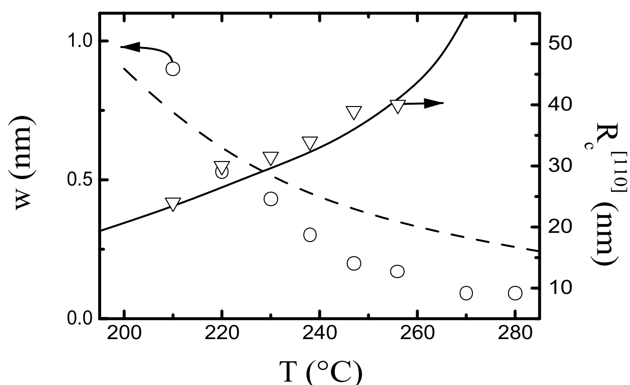

Fig. 3. Surface width w (circles) and mound radius $R_c^{[110]}$ (triangles) as a function of growth temperature for 1 μm thick GaAs epilayers. The dashed and continuous lines represent the simulation results for w and $R_c^{[110]}$, respectively.

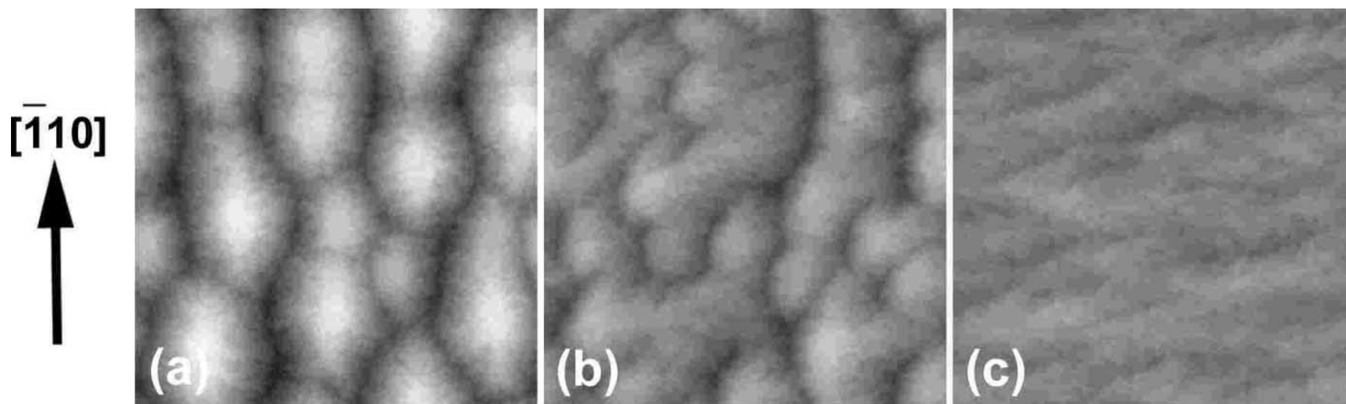

Fig. 4. Gray-scale images of the surface morphologies obtained from growth simulations of 1 μm thick GaAs layers grown at 210 °C on misoriented (001) substrates: (a) 0° (singular); (b) 4° → (111)A; and (c) 8° → (111)A. The area of the images is 200 × 200 nm and the gray scale corresponds to a height difference of 8 nm.

by the ratio of the average separation between 2D islands during deposition of the first layer, σ, to the length of terraces, ℓ; if $\ell < \sigma$ mound formation will be suppressed [3]. To investigate this effect in the case of LT-GaAs and to make an estimate of the misorientation angle ϕ of the vicinal substrate, we performed kinetic Monte-Carlo simulations of the epitaxial growth for several values of ϕ. We choose a (001) substrate misoriented towards the [110] direction (towards the (111)A planes), since the highest surface undulations occur in this direction due to the anisotropic pattern. Surface morphologies obtained from simulating the growth of 1 μm thick layers at 210 °C for different misorientation angles ϕ are depicted in Fig. 4. Fig. 4(a) shows the morphology developed on a singular substrate, where mounds are clearly visible. At $\phi = 4°$, mounds change in shape and become larger in lateral dimensions with a reduced height compared to the singular case. At $\phi = 8°$, the surface becomes significantly smoother, as may be seen from the reduced image contrast. Mound formation has been completely suppressed. The weak features that can be observed on the surface, parallel to [110], are identified as step meandering due to the Bales–Zangwill instability of stepped surfaces [13]. The rms surface width at 0° (singular), 4 and 8° misorientation angle is 0.75, 0.56 and 0.27 nm, respectively. Thus, we conclude that the use of vicinal substrates may reduce significantly the roughness of LT-GaAs epilayers. Preliminary experiments performed on LT-GaAs samples grown on vicinal GaAs(001), 4° → (111)A, confirm these results. Furthermore, these experiments show that epitaxial breakdown occurs at a higher thickness than in the singular case. The results of these experiments will be published elsewhere.

4. Conclusions

We have presented the results of the kinetic Monte-Carlo simulations of the epitaxial growth of LT-GaAs on singular and vicinal substrates. Our model incorporates the deposition and thermal diffusion of adatoms on the substrate. Due to the inclusion of the Ehrlich–Schwoebel barrier at step-edges and an anisotropic diffusion barrier, the simulated surface morphologies on singular substrates and at growth temperatures between 210 and 260 °C exhibit a pattern of elongated growth mounds organized in columns parallel to [$\bar{1}$10]. This pattern is in good agreement with AFM measurements on LT-GaAs samples grown in the same temperature range. Growth on vicinal substrates suppresses the phenomenon of mounding, and at a misorientation angle of 8° the pattern disappears completely while the surface becomes significantly smoother. We propose that the use of vicinal substrates may help to overcome the epitaxial thickness limitations in LT-GaAs.

References

[1] A.L. Barabási, H.E. Stanley, Fractal Concepts in Surface Growth, Cambridge University Press, Cambridge, UK, 1995 (and references therein).
[2] D.C. Look, Thin Solid Films 231 (1993) 61.
[3] M.D. Johnson, C. Orme, A.W. Hunt, D. Graff, J. Sudijono, L.M. Sander, B.G. Orr, Phys. Rev. Lett. 72 (1994) 116.
[4] G. Ehrlich, F.G. Hudda, J. Chem. Phys. 44 (1966) 1039.
[5] R.L. Schwoebel, E.J. Shipsey, J. Appl. Phys. 37 (1966) 3682.
[6] G. Apostolopoulos, J. Herfort, L. Daweritz, K.H. Ploog, M. Luysberg, Phys. Rev. Lett. 84 (2000) 3358.
[7] A. Shen, H. Ohno, Y. Horikoshi, S.P. Guo, Y. Ohno, F. Matsukura, Appl. Surf. Sci. 130 (1998) 382.
[8] J.G. Amar, Fereydoon Family, Phys. Rev. B 54 (1996) 14742.
[9] P. Šmilauer, D.D. Vvedensky, Phys. Rev. B 52 (1995) 14263.
[10] J.W. Evans, D.E. Sanders, P.A. Thiel, A.E. DePristo, Phys. Rev. B 41 (1990) 5410.
[11] D.J. Eaglesham, L.N. Pfeiffer, K.W. West, D.R. Dykaar, Appl. Phys. Lett. 58 (1991) 65.
[12] Z. Liliental-Weber, W. Swider, K.M. Yu, J. Kortright, Appl. Phys. Lett. 58 (1991) 2153.
[13] M. Rost, P. Šmilauer, J. Krug, Surf. Sci. 369 (1996) 393.