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Surface evolution on vicinal GaAs(001) surfaces in the transition range from two-dimensional to step-flow growth

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We have used a 90° double reflection high-energy electron diffraction (RHEED) setup to perform a comprehensive real-time study of the morphology of vicinal GaAs(001) surfaces during molecular beam epitaxy. The technique allows to record RHEED intensities simultaneously in the [110] and [$\bar{1}$10] azimuths and thus enables a detailed study of anisotropy effects. Comparative measurements on surfaces with 2° misorientation towards (111)Ga (A surface) and towards (111)As (B surface), respectively, show that independent on the step type and reconstruction anisotropy, recordings of the specular beam intensity in the azimuth perpendicular to the steps are clearly dominated by the evolution of the staircase order whereas intensity recordings in the azimuth parallel to the steps reveal the evolution of the step edge roughness. Measurements over a wide range of substrate temperatures give insight in the competition between kinetic processes and thermodynamic equilibrium on a length scale accessible to RHEED. For the A surface the transition between two-dimensional (2D) growth and step-flow growth occurs not only at higher temperature than on the B surface, but the disappearance of the intensity oscillations occurs also at different substrate temperatures in different azimuths. The ~20 °C higher disappearance temperature in the [$\bar{1}$10] azimuth is explained with a model based on previous scanning tunneling microscopy results which revealed an increasing elongation of the islands in [$\bar{1}$10] direction with increasing substrate temperature. The B surface is more isotropic and hence no difference in the transition temperature in the two azimuths could be detected. During growth in the transition range between 2D and step-flow growth we observe increased terrace width fluctuations on the B surface, whereas the A surface becomes more uniformly stepped. This demonstrates that in the kinetically controlled regime the anisotropic barrier height for downward diffusion of adatoms over step edges plays an important role for the evolution of the surface morphology. At elevated temperature the barrier height allows downward jumps of the adatoms over B-type steps but not over A-type steps. At conditions close to the thermodynamic equilibrium a kinetic smoothing is observed on the A as well as on the B surface indicating another mechanism to be effective with a change of the energetics due to ordering of the steps in combination with a disordering of the reconstruction on the terraces. This surface is, however, metastable and recovers after growth interruption rapidly (at substrate temperatures >580 °C within less than 1s) to the equilibrium bunched surface. [S0021-8979(97)05006-8]

I. INTRODUCTION

Vicinal surfaces of semiconductors have attracted considerable attention because they are suitable substrates to grow low dimensional structures, such as quantum wires, by molecular beam epitaxy (MBE).[1] For the growth of such structures to be successful, it is necessary to know details of the surface structure and its dynamics under different growth and annealing conditions.[2] In addition, the impact of surface features of non-singular crystal planes on the growth behavior of epitaxial layers and their properties is not fully understood despite a large number of previous studies. To investigate the surface structure of vicinal GaAs(001) surfaces scanning tunneling microscopy (STM),[3–5] atomic force microscopy (AFM),[6] scanning electron microscopy (SEM),[7] reflection high energy electron diffraction (RHEED)[2,5,8–11] and low-energy ion scattering spectroscopy (LEIS)[12] were used. STM investigations revealed a quite different step structure for the vicinal GaAs(001) surface misoriented towards [$\bar{1}$10] (hereafter called the A surface) and the vicinal GaAs(001) surface misoriented towards [110] (B surface), respectively. The Ga-terminated steps (A-type steps) on the A surface are much smoother compared to the As terminated steps (B type steps) on the B surface, but they show a less homogeneous terrace width distribution.[3] This confirmed earlier conclusions from profile measurements of the specular RHEED beam.[8] Similar information was obtained from low energy ion scattering experiments where the amount of backscattered He as function of the incidence direction is very sensitive to the step structure in the direction of the incident beam.[12]

STM investigations after quenching of growing surfaces showed agglomerates forming islands that are elongated in the [$\bar{1}$10] direction.[4,5,13–15] It has to be realized, however, that with increasing growth temperature the problem to properly freeze in the structure of the growing surface for STM investigations becomes more crucial. A powerful tool to de-

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termine the surface structure *in situ* is RHEED. Although it is an averaging method and the real growth process—in contrast to established simple growth models—can be rather complicated and inhomogeneous as recently found by *in situ* SEM studies,[7] RHEED is capable to provide information about the surface structure in a number of ways.

Analyzing the profile of the specular RHEED beam provides insight into step roughness and terrace width distribution.[8,9,16] If the incident beam is directed down the staircase, information about the terrace uniformity is obtained whereas in the azimuths parallel to the steps the step roughness can be assessed. Besides the type of the steps, the surface reconstruction and growth conditions were found to be important in establishing the staircase order.[8] The step edge structure depends on the substrate temperature during growth. For growing surfaces a smoothing of the step edges was reported to occur with increasing substrate temperature (the width of the specular beam decreased), whereas without Ga supply roughening of the surface was observed with increasing substrate temperature.[17] This indicates a rather complex interplay between thermodynamics and kinetics, as the adsorption and desorption rates are substrate temperature dependent.

The change of intensity of the specular RHEED beam after start and termination of growth is related to different processes. The recovery process of the specular intensity is described as a combination of an initial fast recovery and a slow process.[18] For the (2×4) reconstructed surface it has been proposed that the initial recovery involves rearrangement of the terrace configuration due to surface diffusion of Ga atoms, whereas the second step involves the restoration of long-range order with breaking and formation of chemical bonds. It is noteworthy that the time constant of the recovery rate of the specular beam intensity decreases in the temperature range between 515 and 590 °C by more than one order of magnitude.[19]

Measurements of RHEED intensity oscillations of the specular beam during growth at different substrate temperatures revealed that a transition of the growth mode takes place at a critical substrate temperature which depends on the step type.[2,10] Below that transition temperature, the growth process is dominated by two-dimensional nucleation and island growth. Increasing the substrate temperature above the transition temperature changes the growth mode to step flow growth. Recently it has been shown by RHEED,[20] STM,[21] and SEM[7] that the two growth modi can coexist on a terrace with nucleation at the terrace centre accompanied by step propagation. Therefore, a dynamical transition between the two growth modi rather than an abrupt transition is expected. It is described as a change in the relative probabilities to nucleate a new island versus attaching atoms to an existing step edge.

RHEED intensity studies are usually performed with the electron beam incident parallel to the misorientation steps. This provides in principle one-dimensional information. The structure of GaAs(001), and in particular of the vicinal surfaces is, however, highly anisotropic. The behavior of the atomic species on the surface during growth is therefore likely to be anisotropic in some respect. The investigation of the Ga adatom diffusion on the As-stabilized GaAs(001) surface by first-principles pseudopotential methods revealed an anisotropy factor of ~3 between the activation barrier for diffusion along [110] and $[\bar{1}10]$, respectively.[22] This is consistent with RHEED measurements on vicinal surfaces from which it was concluded that the surface diffusion length for Ga is strongly dependent on the experimental conditions but may be up to one order of magnitude larger in $[\bar{1}10]$ than in [110] direction.[23] It is important to note that the chemical nature of steps in the $[\bar{1}10]$ direction is completely different from that in the [110] direction and that the RHEED experiment cannot separate the diffusion characteristics and the characteristics of the step sites.[24] The comparison of RHEED measurements with Monte Carlo growth simulations led to the conclusion that the origin of the anisotropic growth-mode transitions stems mainly from anisotropic incorporation kinetics rather than from anisotropic adatom mobility.[25] Both experiments and theoretical considerations suggest that the probability of sticking on B-type steps is higher than on A-type steps.[13,26]

Keeping in mind the two-dimensional character of the surface, monitoring the intensity of the specular RHEED beam in more than one direction is supposed to provide information about the nature of this anisotropic behavior as well as about the interplay between thermodynamic and kinetic processes on the growing surface. This objective can be achieved with the time synchronized double RHEED experiments. A first investigation into simultaneous RHEED intensity oscillation behavior was carried out—to our knowledge for the first time—by using an experimental setup using two electron guns.[27] The advantage of the double RHEED setup is that the RHEED pattern in two azimuths is obtained at the same substrate temperature from the same substrate location. From a practical point of view it is desirable to get a more detailed understanding of the microstructural anisotropy because macroscopic properties such as Hall mobilities also show anisotropic behavior.[28]

In this work we apply the double RHEED technique to investigate anisotropy effects during GaAs growth and annealing on A and B surfaces. The outline of this article is as follows. First we describe the experiment with the simultaneous observation of the RHEED patterns in two nonequivalent $\langle110\rangle$ azimuths. Then the intensity behavior during growth and after growth interruptions is discussed in terms of the competition between kinetic processes and thermodynamic equilibrium as a function of substrate temperature. To explain characteristic differences in the time behavior of the RHEED intensity for A and B surfaces, we consider an anisotropic barrier for downward diffusion of adatoms over A- and B-type steps. From the azimuthal dependence of the disappearance temperature of RHEED intensity oscillations we draw conclusions on the anisotropy of surface processes and of step edge structures as well as on the dynamical transition between two-dimensional and step-flow growth. Finally the relation between RHEED and STM/AFM results is discussed.

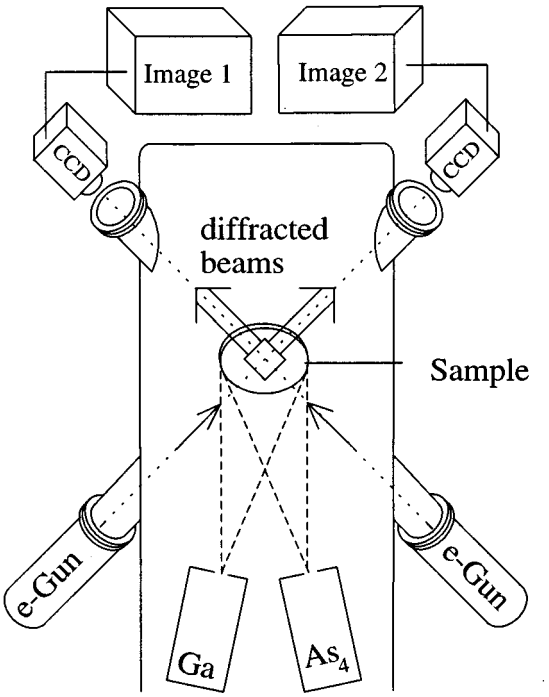

FIG. 1. Experimental setup for double RHEED experiments.

II. EXPERIMENTS

As shown in Fig. 1, two electron guns were installed in a specially designed MBE system (MECA 2000) to observe the RHEED pattern of the sample in two perpendicular, i.e., the [110] and [1$\bar{1}$0] azimuths. The chamber is constructed in such a way that the both electron beams intersect on the sample at an angle of 90°. The substrate is positioned in the cross-point and rotated until the electron beams are parallel to the [110] and [1$\bar{1}$0] crystallographic directions of the substrate. For MBE growth the system was equipped with Ga dual filament and As_4 effusion cells. Epiready GaAs(001) wafers with misorientation of 2° towards (111)Ga or 2° towards (111)As were used as substrates. The base pressure prior to the experiment was in the 10^{-11} Torr range. The growth rate was 0.7 ML s^{-1} in all experiments. To achieve a high sensitivity during image acquisition of both specular beams, a very small incidence angle of the electrons of approximately 0.4° was chosen in the directions parallel as well as perpendicular to the steps. The energy of the electrons was 20 keV. The substrate temperature was measured with a thermocouple pressed to the back side of the substrate. The temperature of the sample specimen was calibrated to the oxide desorption temperature of GaAs.[29] To diminish the influence of the substrate temperature gradient and to bring the two electron beams to the same area on the sample, we used small substrate pieces of 10×10 mm^2. The accuracy of temperature differences for experiments carried out on the same sample is supposed to be better than 5 °C, whereas the accuracy of the absolute temperature estimate concerning different samples has an error margin of up to 20 °C, since the oxide desorption temperature depends slightly on the history of the wafer.

In order to avoid systematic structural changes, experiments at different substrate temperatures were not carried out in a sequence with monotonous increase or decrease of the substrate temperatures. Higher and lower substrate temperatures were chosen at random. Returning to a previous substrate temperature after a couple of experiments showed no significant change in the oscillation behaviour. The ratio of the beam equivalent pressures (BEP) As_4/Ga estimated to be 15 before growth at the sample position was kept constant in all the experiments to maintain a (2×4) surface reconstruction during growth and to ensure step flow growth on the vicinal (001)B surfaces. After each experimental run the surface was allowed to recover under an As_4 flux at a substrate temperature of 580 °C for a few minutes.[2,15]

The intensity of the specular beam was monitored in both azimuths with two charge coupled device (CCD) cameras connected to two independent image processing systems (Fig. 1). The diffraction patterns were recorded on videotape. The signals of light emitting diodes (LED) overlaid on the RHEED pattern were used to indicate the shutter status and to align the intensity curves on a time scale. In our experiments the RHEED intensity oscillations for A and B surfaces with 2° misorientation were always measured simultaneously in the exact [110] and [1$\bar{1}$0] azimuths. The oscillation curves shown in Figs. 2 and 3, respectively, represent as-measured curves without any smoothing procedure.

III. RESULTS AND DISCUSSION

To clarify the information that can be extracted from recordings of the specular RHEED beam intensity with the electron beam parallel and perpendicular to the steps, we first compare the RHEED intensity behaviour in the [110] and [1$\bar{1}$0] azimuths for A and B surfaces. The competition between kinetic processes and thermodynamic equilibrium is illustrated by means of measurements over a wide range of substrate temperatures. In particular, we demonstrate the role of an anisotropic barrier height for downward diffusion of adatoms over step edges in the kinetically dominated regime and rapid changes in morphology between the static and dynamic surface at conditions close to the thermodynamic equilibrium. Then differences between A and B surfaces concerning the azimuthal dependence of the disappearance temperature of RHEED intensity oscillations are discussed within a model that includes island formation and step edge roughening due to the anisotropy in adatom migration as well as in step edge incorporation.

A. Competition between kinetic processes and thermodynamic equilibrium

Figures 2 and 3 show recordings of the specular beam RHEED intensity for GaAs growth on the A and B surfaces, respectively, that were measured simultaneously in the [110] and [1$\bar{1}$0] azimuths. Their comparison allows us to check whether the measured intensity curves are directly correlated with changes in the density, distribution, and shape of the atomic steps. Since the intensity will be mainly affected by the step component perpendicular to the direction of the incident beam, the recordings in the azimuths parallel and perpendicular to the nominal step direction, respectively, are

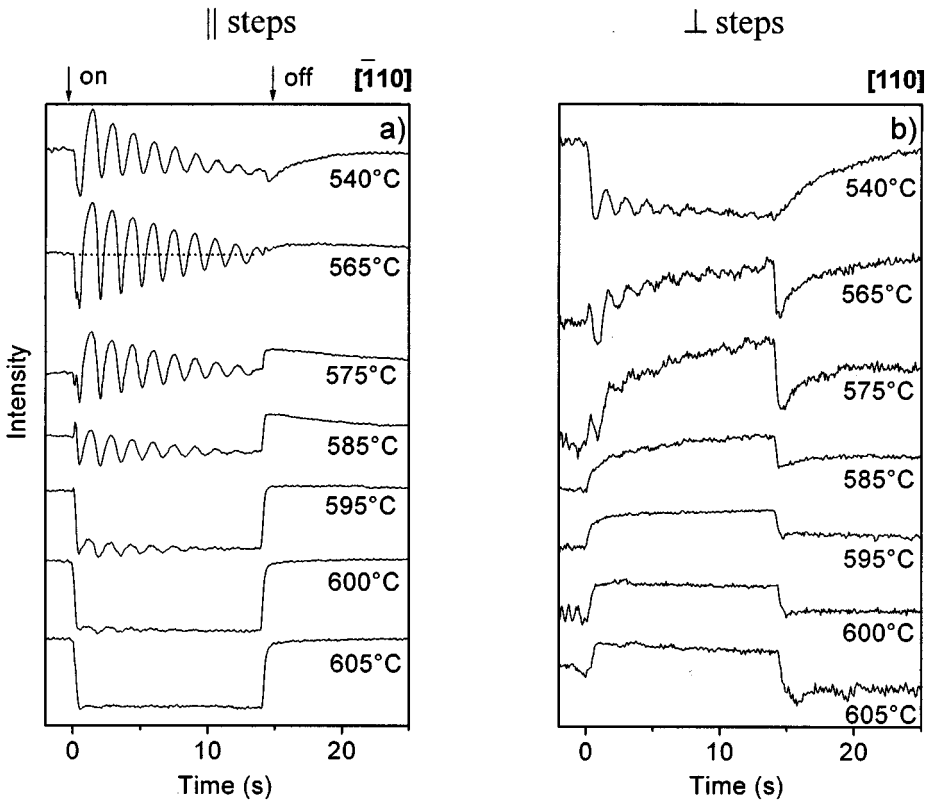

FIG. 2. Intensity of the specular beam during and after growth on a vicinal (001)A surface for different substrate temperatures as a function of time, recorded simultaneously in the (a) [1̄10] azimuth and (b) [110] azimuth.

expected to be sensitive to step edge roughness and terrace width fluctuations, respectively.[16] At high substrate temperatures where the growth proceeds completely in the step flow mode and therefore no intensity oscillations are visible, a striking difference is observed between the intensity behavior in the two azimuths. For the A surface the intensity decreases during growth in the [1̄10] azimuth, whereas it increases in the [110] azimuth [Figs. 2(a) and 2(b)]. The opposite behavior is found for the B surface [Figs. 3(a) and 3(b)], where the intensity decreases in the [110] azimuth and increases in the [1̄10] azimuth. The vicinal A and B surfaces show, however, a similar intensity behavior for the direction parallel to the steps [compare Figs. 2(a) and 3(a)] and for the direction perpendicular to the steps [compare Figs. 2(b) and 3(b)]. This clearly indicates that the intensity behavior is caused or at least dominated by the evolution of the step structure. If the change in the specular intensity would be due to changes in surface reconstruction (either symmetry or degree of order) one should expect a similar intensity behavior in the same azimuths of the different vicinal surfaces. This is not observed.

The symmetry relation of the intensity behavior with respect to the ⟨110⟩ directions found at high temperatures holds less pronounced also in the low temperature range [540 °C for the vicinal (001)A surface, 510 °C for the vicinal (001)B surface] where clear oscillations prove two-dimensional nucleation (Figs. 2 and 3). This is expected because now islands elongated in the [1̄10] direction on both vicinal surfaces are formed on the staircase-like structure of the vicinal surface. The more complicated RHEED intensity behavior in the transition range between 2D and step flow growth will be discussed in the next paragraph for the two types of vicinal surfaces.

To further examine a possible influence of changes in surface reconstruction on the behavior of the specular beam intensity we have compared simultaneous intensity recordings of the specular beam and of fractional-order beams for the B surface. Figure 4 shows recordings of the half-order beam in the [110] azimuth and of the quarter-order beam in the [1̄10] azimuth at a substrate temperature of 545 °C and 580 °C, respectively. At 580 °C growth initiation leads to a relatively strong decrease in the As coverage of the (2×4) reconstructed surface. This transition of the (2×4) reconstructed surface to a lower As coverage at 580 °C after growth initiation leads to a drastically reduced intensity of the fractional-order reflections in both azimuths. The specular beam intensity decreases in the [110] azimuth and increases in the [1̄10] azimuth suggesting that it is not directly coupled with the fractional-order beams. It can, however, not be excluded that a change in the structure near the step edge plays a role.

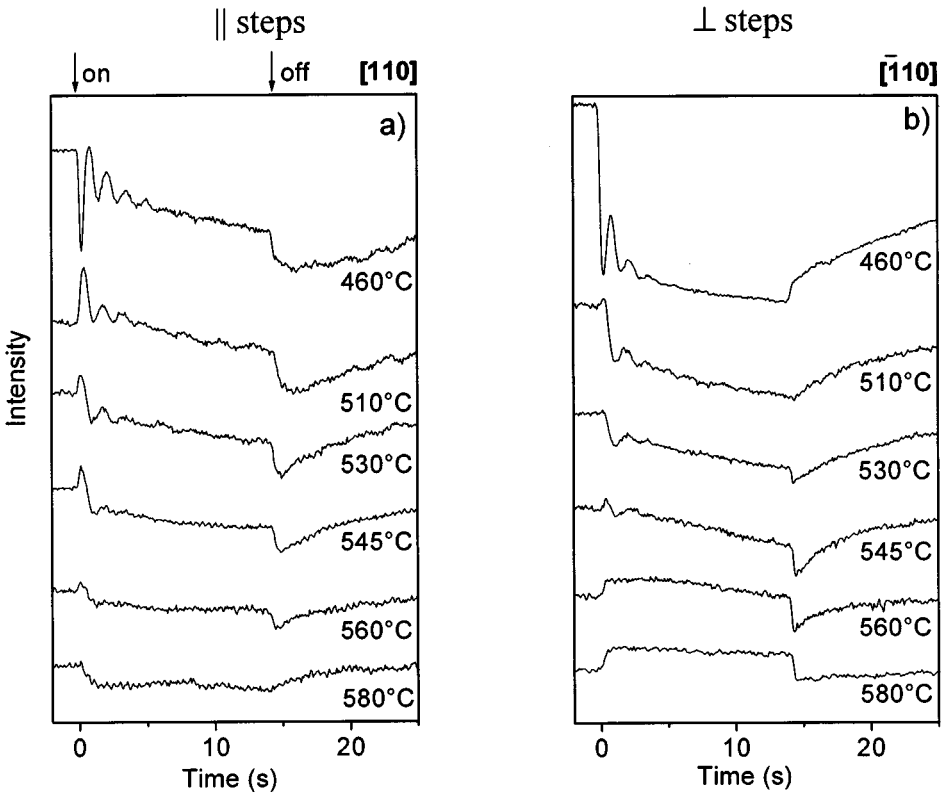

FIG. 3. Intensity of the specular beam during and after growth on a vicinal (001)B surface for different substrate temperatures as a function of time, recorded simultaneously in the (a) [110] azimuth and (b) [$\bar{1}$10] azimuth.

The measurement at an intermediate substrate temperature of 545 °C is of interest because of the intensity overshoot and undershoot phenomena observed when switching between the static and the growing surface. Since the intensity recordings of the fractional-order beams at 545 °C do not show intensity overshoot or undershoot phenomena in comparison to the specular beam the corresponding phenomena have to be ascribed to changes in the step structure rather than in surface reconstruction. After growth interruption, the intensity of the specular beam (Figs. 2 and 3) shows the well known recovery behavior.[11] It is noteworthy that in case of the A surface the intensity in the azimuth perpendicular to the steps can even exceed the initial value. Above a substrate temperature of about 580 °C there is virtually no delay to reach the pregrowth intensity (less than 1 s) for both vicinal surfaces in both azimuths. The intensity recovery process was described as a combination of a rapid initial recovery (activation energy 1.5 eV) and a slow process (activation energy 5 eV).[18] Neave *et al.*[19] estimated a decrease in the time constant for the recovery of the specular beam intensity by more than one order of magnitude in the temperature range 515–590 °C. The present observations agree qualitatively with those results. From the discussion above we conclude that both the fast and the slow process are due to ordering processes that are related to adatom migration and/or changes in the step structure and step width distribution.

At a substrate temperature of about 580 °C for the B surface and about 600 °C for the A surface, the RHEED intensity reaches constant values for the nongrowing and growing surfaces, respectively, within less than 1 s. This proves that the kinetic processes are rapid so that the system can approach the thermodynamic equilibrium on the length scale probed by RHEED. Even after loss of the oscillations some conclusions on surface dynamics can be drawn from the rapidity and amount of intensity changes after growth initiation/interruption and from the observation whether constant intensity levels are reached. In particular for the A surface a rapid switch between (nearly) constant, rather different intensity levels is observed in the azimuth parallel to the steps where the intensity is sensitive to step edge roughness and adatom concentration. Since A-type steps are relatively smooth, it is concluded that this intensity behavior is dominated by changes in adatom density. This suggests that above the (apparent) transition to step flow growth, which will be discussed later in detail, a high Ga adatom concentration exists on the terraces that changes rapidly after growth interruption. Kinetic as well as thermodynamic reasons might be responsible for this. Both the higher activation barrier for diffusion along [110] as compared to that along [$\bar{1}$10][22] and a lower probability for sticking at A-type steps compared to that on B-type steps[13,26] can lead to a high adatom concentration. A surprising high adatom concentration can exist,

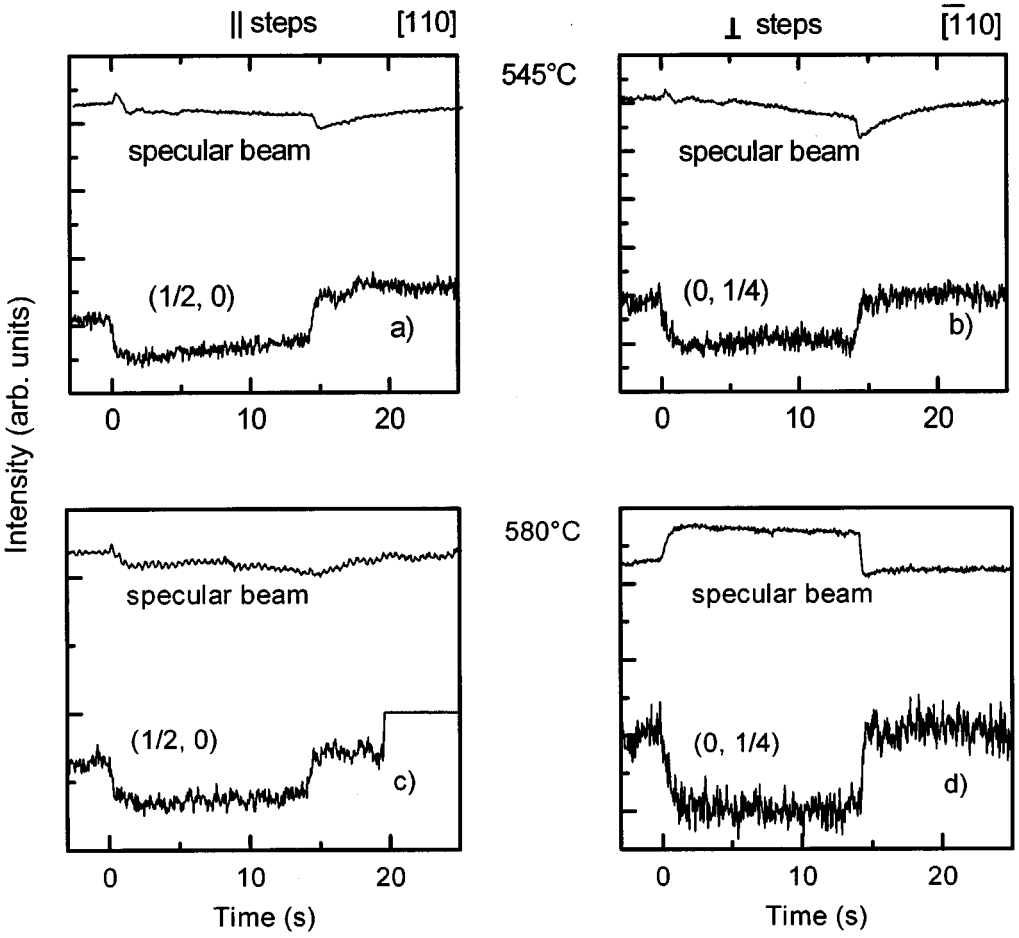

FIG. 4. Intensity of the specular beam and the (1/2,0) and (0,1/4), respectively, superstructure reflexes during and after growth on the vicinal (001)B surface at a substrate temperature of (a) 545 °C and (b) 580 °C.

however, already during equilibrium. In a recent STM study[30] the thermal concentration of adatoms for typical MBE growth temperatures and As overpressures was frozen into small islands on the terraces by rapid cooling and estimated from measuring the area of the islands. For a 0.15° misoriented surface, e.g., a concentration as high as 0.18 ML at 600 °C was found. As a consequence of these results we conclude that in the high temperature range besides kinetic effects the equilibrium concentration is an important factor for reordering processes between the dynamic and static surface. The adatom concentration will differ not only simply by the amount corresponding to the incoming flux but in addition also due to a shift in the equilibrium adatom concentration which is related to a change in surface stoichiometry. In analogy to considerations for the static surface[30] we speculate that at less As-rich conditions (during growth) the equilibrium Ga adatom concentration increases.

Below 580 °C for the B surface and 600 °C for the A surface the competition between kinetic processes and the system's intention to reach the thermodynamic equilibrium is clearly shifted in favor of the kinetics. In this temperature range a striking difference in the intensity behavior during growth is observed between A and B surfaces. For the A surface, the average intensity as defined in Fig. 2(a) as a dotted line, is approximately constant in the azimuth parallel to the steps. In the perpendicular azimuth, however, it increases for temperatures above 540 °C and below 600 °C. In the case of the B surface the average intensity decreases continuously during growth in both azimuths, down to the lowest investigated temperature of 460 °C (Fig. 3). The opposite behavior of A and B surfaces in the azimuth perpendicular to the steps can be understood within models considering the barrier for downward diffusion of adatoms over steps.[31–33] If the barrier is large or infinite for downward flow the preferential adatom attachment to the upper step edge will lead to a terrace width equalization, because the step velocity is directly proportional to the size of the feeding range. In the opposite case of a small barrier, atoms arriving on the terrace can approach the step with a higher probability from the upper terrace and hence the terrace width fluctuation will increase. Supposing that the ordered step array leads to a higher specular beam RHEED intensity, the observed intensity behavior suggests a terrace width equalization on the A surface and an increased terrace width fluctuation on the vicinal B surface. Therefore, we conclude that the diffusion over the steps is anisotropic with a considerably higher barrier for the A-type steps than for the B-type steps. This is consistent with recent STM results on the

growth front morphology of the singular GaAs(001) surface.[13] There the surface consists of a multilevel system of terraces occupying up to seven levels in the [110] direction, whereas along [110] only one level is typically visible over a distance of several thousand Å. Downward transport over steps is very likely since otherwise, in contrast to the observations, the growth front roughness would increase very rapidly during growth in nonstep-flow growth modes. The experiments of Heller and Lagally[13] also indicated a barrier for transport at least over A-type steps.

B. Azimuthal dependence of the disappearance temperature of RHEED intensity oscillations

1. Vicinal GaAs(001) A surface

To discuss the behavior of the specular intensity measured simultaneously in different azimuths it is useful to consider the origin of the oscillations. According to the established model the 2D growth proceeds as nucleation, growth and coalescence of islands.[7,10] As shown by STM investigations, the islands are elongated along the [110] direction.[4,5,13–15] Combined RHEED and STM experiments were carried out to correlate the specular beam intensity and the step length per area.[4] Those experiments were carried out on a rather rough time scale and the nucleation stage cannot be sufficiently explained on the basis of STM images taken after sample quenching. Using the results of simultaneous RHEED intensity recordings in two azimuths we get additional information on the evolution of the step array and can verify the general assumption that the relevant step edge to explain the intensity behavior is the component perpendicular to the incident electron beam.

Figure 2 shows the intensity behavior of the specular beam for the A surface at different substrate temperatures. In the [110] azimuth parallel to the step edges the transition temperature from 2D to step flow growth is 605 °C [Fig. 2(a)] as indicated by the disappearance of the oscillations. In the [110] azimuth, perpendicular to the steps, the intensity shows a considerably enhanced noise level [Fig. 2(b)] indicating a less ordered structure detected in this direction. Nevertheless, the intensity oscillations are clearly visible and a transition temperature of 585 °C between the two growth modi is estimated. The remarkable difference between the transition temperatures in the two azimuths was found to be reproducible in different experiments. Due to the noise there is an uncertainty in estimating the exact disappearance temperature of the oscillations. Since this error does not exceed 5 °C and the transition temperature difference was reproducibly found to be larger than this margin, this difference is certainly an effect caused by the surface structure.

The fact that on the A-surface intensity oscillations disappear in the [110] azimuth at a 20 °C lower substrate temperature than in the [110] azimuth can be explained in terms of the changing shape of the growing island when the transition temperature is approached. Following the assumption that the component of the step length per area normal to the electron beam is mainly responsible for the oscillations in this azimuth, one has to conclude that the step length per area reaches its steady state value in the [110] azimuth at a lower substrate temperature than in the [110] azimuth. This is consistent with the simple model presented in Fig. 5 which is based on the well established tendency of the (2×4) reconstructed GaAs(001) surface to form islands elongated in the [110] direction.

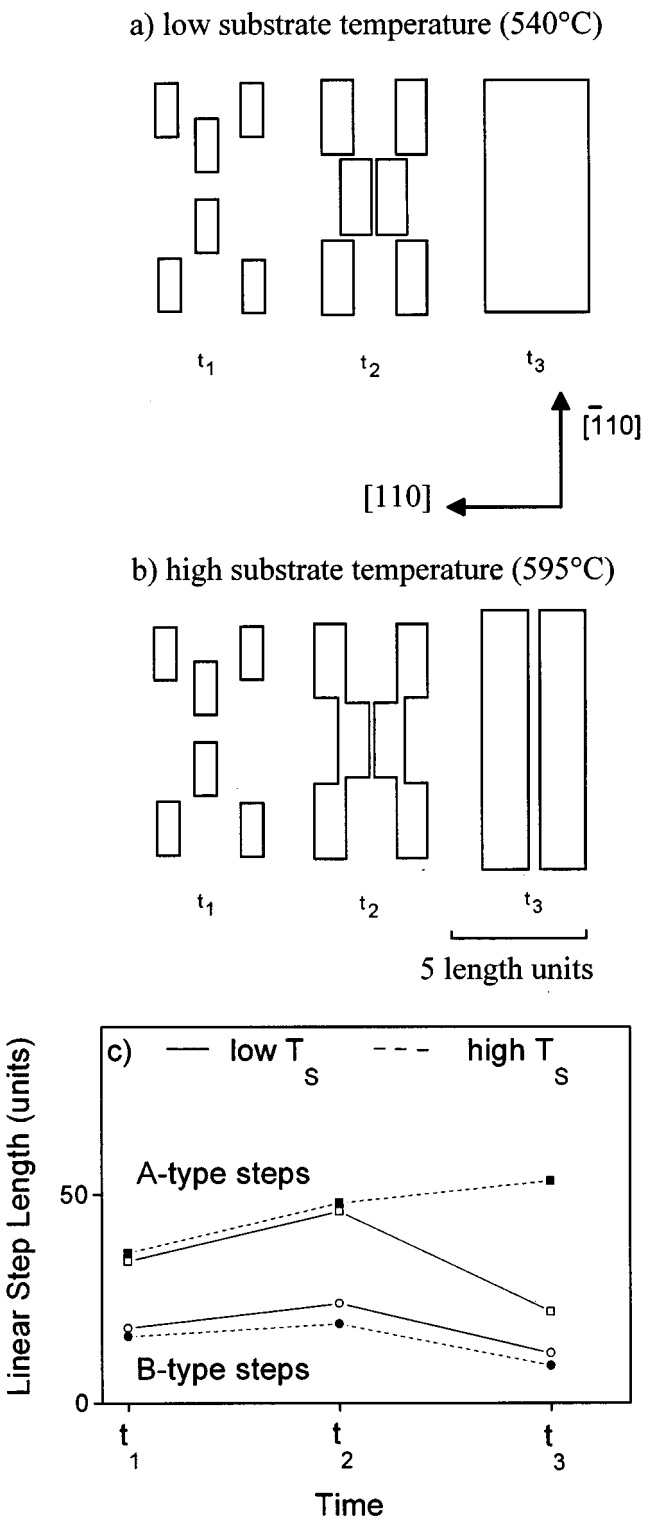

FIG. 5. Schematic illustration of development of island shape for different substrate temperatures on a terrace of a vicinal (001)A surface. The same surface area is considered at times t_1, t_2 and t_3: (a) lower substrate temperature, (b) higher substrate temperature, (c) linear step length for cases (a) and (b).

To illustrate the main idea, the model neglects that in reality the islands may have different sizes and their shape is not rectangular but ragged. The higher substrate temperature enables the Ga atoms to diffuse more easily thus enhancing the probability that they reach a step and stick there. Since the migration proceeds preferably along [110] and the probability of sticking on B-type steps is higher than on A-type steps, the islands are growing anisotropically in length. At lower substrate temperatures [Fig. 5(a)] the islands are elongated as well. But they more or less preserve their axis ratio during growth. This leads to a periodically changing step length per area in both the [110] and [110] azimuths [Fig. 5(c)]. Therefore, intensity oscillations can be observed in both azimuths. For enhanced substrate temperature [Fig. 5(b)] it is important to note that the island elongation is not solely influenced by the factors already discussed. The tendency to anisotropic growth will be even more increased by the downward diffusion over B-type steps that has been discussed in the Sec. III A. The islands are getting larger during growth with the length of A-type steps approaching a steady state value, whereas the B-type steps are changing their length per unit area periodically as a function of time [Fig. 5(b)]. Accordingly, intensity oscillations occur in the [110] azimuth but cannot be observed in the [110] azimuth any longer [Fig. 5(c)]. These results are consistent with SEM observations[7] that step flow and 2D growth occur simultaneously and conclusions from STM work[21] that there is a dynamical transition between the two growth modi rather than an abrupt change.

2. Vicinal GaAs(001)B surface

Figure 3 shows the RHEED intensity behavior of the specular beam for the B surface at different substrate temperatures. At low temperatures oscillations are clearly visible in the [110] and [110] azimuths although they are less pronounced compared with the A surface. The higher degree of disorder on the B surface is supposed to make the detection of the intensity oscillations more difficult. The strong meandering of the As-terminated steps during growth is evident from the continuously decreasing intensity in the [110] azimuth (parallel to the steps) where the intensity is sensitive to step-edge roughness. It is noteworthy that there is a simultaneous decrease of the specular beam intensity in the [110] azimuth (perpendicular to the steps), indicating that the step meandering is coupled with an increase in terrace width fluctuation.

At elevated substrate temperatures the RHEED signal-to-noise ratio deteriorates. Under the assumption of at least two distinguishable intensity maxima to be present the temperature for the transition from 2D to step flow growth is estimated to be 560 °C for both the [110] azimuth (parallel to the steps) and the [110] azimuth (perpendicular to the steps). In several experiments we could not find a difference between the transition temperatures for the two azimuths which exceeded the error margin of 5 °C. This is in clear contrast to the vicinal (001)A surface. It is obvious that due to step meandering the vicinal (001)B surface is more isotropic regarding surface roughness and consequently also regarding the substrate temperature at which the RHEED intensity oscillations disappear.

The surface evolution below the transition to complete step flow can be well explained by the preferential adatom migration along the [110] direction, the higher sticking probability of Ga atoms at B-type steps, and a high probability of an adatom crossing downward B-type steps. Above the transition to entire step flow ($\geqslant$560 °C), the behavior of the RHEED intensity in the [110] azimuth indicates that after growth initiation the step edge roughness increases but reaches a steady state value within about 1 s [Fig. 3(a)]. Surprisingly, the rapid increase of the RHEED intensity after growth initiation to a constant value in the [110] azimuth suggests a ''kinetic smoothing'' due to terrace width equalization. Therefore, in this temperature range another mechanism than adatom crossing downward B-type steps must be effective. We have to consider that in the competition between kinetic effects from the arriving atoms onto the surface and surface tension forces which drive the structure back toward equilibrium,[34] the latter now becomes dominant. It is important to note that in this limited temperature range the fractional-order beams disappear during growth (Fig. 4), indicating that the ordering (unbunching) of the steps and disordering of the reconstruction on the terraces are linked and that the energetics on the resulting metastable surface is significantly changed. It is the same mechanism that becomes operative on vicinal Si(111) in a narrow temperature range beneath the equilibrium faceting temperature.[34] In this case the kinetic smoothing of the equilibrium faceted surface morphology during Si growth is accompanied by the disappearance of the (7×7) reconstruction. In contrast to the metastable smoothed vicinal Si(111) surface which is remarkably robust, the kinetically smoothed vicinal GaAs(001)B surface recovers to the equilibrium bunched surface within less than 1 s.

C. Relation between RHEED and STM/AFM results

RHEED and STM are complementary methods. RHEED averages over the illuminated sample area, whereas STM makes a detailed real-space study of much smaller surface areas possible. From the practical point of view, the main differences in sample handling between STM and RHEED are that in STM the investigations are performed after quenching and transferring of samples whereas the RHEED intensity characterises a sample area several order of magnitude larger *in situ*. The present study has shown that at substrate temperatures of 580 °C and above, the recovery time between the structure of the dynamic and the static surface is less than 1 s (Figs. 2 and 3, respectively). During this time dramatic changes in the surface morphology occur. This imposes crucial problems to quench the actual surfaces for STM studies. On the other side, the time-dependent RHEED intensity recording gives insight into the surface dynamics but cannot supply us with quantitative data on island shape and size. Moreover, the usual recording in only one azimuth ignores in principle the two-dimensional character of the surface structure and processes. It is therefore challenging to relate results of experiments carried out in the double RHEED geometry to results from STM investigations.

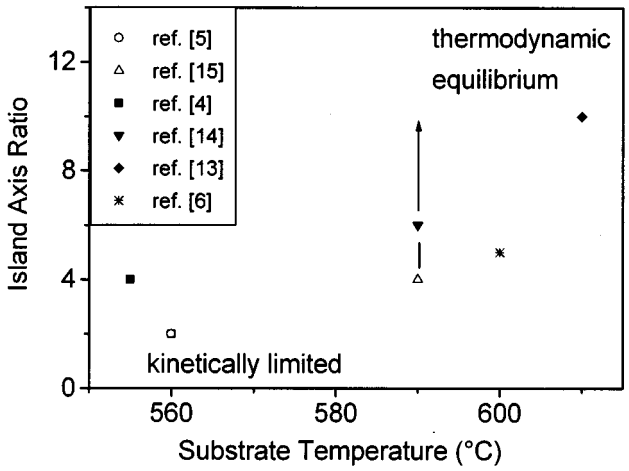

FIG. 6. Axis ratio of GaAs islands on nominally singular GaAs(001) surfaces (solid symbols) and on a vicinal GaAs(001)B surface (open symbol) for different substrate temperatures.

From the intensity oscillation behavior it has been concluded that the island elongation along [110] increases in the 2D growth regime with substrate temperature (see model in Fig. 5), not only due to anisotropy in adatom migration and attachment on A-type and B-type steps but in particular due to downward diffusion over B-type steps. The transition in the growth mode from 2D to step flow is completed at about 605 °C, i.e., in a temperature range for which we expect near-equilibrium conditions. To verify this, Fig. 6 summarizes data on the axis ratios of islands derived from STM and AFM observations by several authors for nominal singular (001) surfaces (solid symbols) and for the vicinal (001)B surface (open symbol). If not given by the authors, we have estimated the axis ratio from the smaller islands. At substrate temperatures $\leqslant$590 °C there is a tendency that the axis ratio is smaller for islands near B-type steps as compared to islands on the singular surface (or near A-type steps). This can be explained by the preferential migration along the [110] direction leading to differences in the size of the feeding areas near A- and B-type steps. At a substrate temperature of 590 °C the island axis ratio increases from $\sim$4 for rapidly cooled samples[4] up to $\sim$10 for annealed samples (indicated by arrows).[15] A shape anisotropy of $\sim$10:1 has been found also for sample growth and 15 min annealing at 610 °C,[13] suggesting that this value represents the equilibrium anisotropy. This agrees well with our conclusion that the island shape anisotropy increases with temperature until a constant value is reached at near-equilibrium conditions.

D. Conclusions

The surface morphology on vicinal GaAs(001) surfaces during and after MBE growth has been studied in real-time with a 90° double RHEED setup. This technique allows to record the intensity behavior of the specular and fractional-order beams simultaneously in the orthogonal [110] and [110] azimuths and to perform a two-dimensional analysis of the evolution of the surface structure and of the underlying atomic processes in a wide range of substrate temperatures.

On the A surface the disappearance temperature of the RHEED intensity oscillations is about 20 °C higher in the [110] azimuth than in the [110] azimuth. This is due to the formation of elongated islands which change their axis ratio in the transition range between 2D and step flow growth as a function substrate temperature. As a consequence of the strong meandering of As-terminated steps, the B surface is more isotropic and no measurable difference between the disappearance temperatures of the RHEED intensity oscillations in the two orthogonal $\langle 110 \rangle$ azimuths is found. During growth in the transition range between the 2D and step flow mode the A surface becomes more uniformly stepped, whereas on the B surface an increase of the terrace width fluctuations is observed. This is explained by an asymmetry in the step attachment with a downward diffusion of adatoms over B-type steps. At near-equilibrium conditions above $\sim$580 °C, a kinetic smoothing occurs also on the B surface. In this case the terrace width equalization is linked with a disorder of the reconstruction and therefore not the result of the step attachment asymmetry but rather a consequence of the energetics. The unbunched B surface is metastable and recovers after growth interruption rapidly to the equilibrium bunched surface. At high substrate temperatures the bunched surface is also the equilibrium state of the A surface. This finding is consistent with conclusions from STM[14,15,35] and AFM observations.[6]

The results demonstrate that the 90° double RHEED technique can be advantageously used to study metastable surfaces that are difficult to quench in for STM studies. But even in this case the imaging in real space by STM is valuable for the detailed interpretation of the RHEED results.

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