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Terrace width distribution during unstable homoepitaxial growth of GaAs(110): An experimental study

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Abstract

The temporal evolution of the step bunching instability formed during GaAs homoepitaxial growth on the GaAs(110) vicinal to (111)A has been studied by atomic force microscopy (AFM) and the step–step distribution has been quantified as a function of deposition time. Analysis of the AFM data has shown that neither the terrace width distribution (TWD) nor the terrace height distribution (THD) fit to a Gaussian function in the initial stages of growth, but both evolve with time as the bunching instability develops. After deposition of 500 ML of GaAs the TWD exhibits a clear Gaussian behavior while the THD is very well fitted to a Lorentzian distribution. The GaAs surface morphology initially shows a great dispersion in terrace height and width values with a clear anisotropy along the <001> tilt direction, but evidence of self-controlled growth is observed irrespective of layer thickness.

Keywords: Homoepitaxial growth vicinal GaAs(110); Temporal evolution step bunching; Terrace width distribution

1. Introduction

Unstable growth of GaAs by molecular beam epitaxy (MBE) on (110) vicinal substrates offers a new and attractive way to fabricate laterally ordered nanostructures [1,2]. A prerequisite to obtain such structures is to have a detailed understanding of the mechanisms that govern the destabilization of the growth front during homoepitaxy. Depending on the experimental conditions, one or various morphological instabilities derived from the presence of Ehrlich–Schwoebel barriers at step edges can arise on this surface during growth in the step flow mode. Basically, these are the so-called step meandering formed by the Bales–Zangwill mechanism [3] and the step bunching instability [4,5]. Specifically, the step bunching instability is formed during growth on a vicinal crystal surface when a regular train of equally spaced steps decomposes into alternating low step density regions (terraces with orientation close to a singular surface) and high step density regions (bunches). The process can be driven energetically by an attractive step–step interaction, or by a variety of kinetic mechanisms. In the latter case, step bunching is caused by breaking the adatom incorporation symmetry between the ascending (upper) and descending (lower) steps. Preferential attachment to the upper step edge tends to equalize terrace lengths, while attachment to the lower step edge destroys terrace width uniformity and leads to step bunching [6,7]. Therefore, the terrace width distribution (TWD), which gives the probability of finding a terrace of width l on the surface, provides valuable information regarding the step–step interactions during growth.

The formation of step bunches on the GaAs(110) surface has been attributed to a preferential incorporation of Ga adatoms to steps from the upper terraces, which typically occurs during growth in the As-rich/Ga supply-limited regime, that is, at low growth rates and relatively low temperatures. The GaAs growth rate, under such conditions is kinetically controlled by the arrival rate of Ga to the surface [8]. We have recently corroborated the intrinsic origin of this instability during atomic hydrogen-assisted oxide desorption experiments prior to MBE growth [9]. To our knowledge, a detailed experimental study of the terrace width distribution during step bunching on the GaAs(110) vicinal surface has never been carried out in the past, although a number of general theoretical

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studies can be found in the literature [10–12]. In this article, we present a quantitative AFM study of the temporal evolution of the terrace width distribution during MBE growth of GaAs (110) in the Ga supply-limited regime with a view to understanding the step–step interaction kinetics that lead to step bunching formation.

2. Experimental details

The experiments were carried out in a standard Varian-360 MBE system equipped with a 10 keV electron gun for reflection high-energy electron diffraction (RHEED) measurements. Ga and As_4 fluxes were calibrated using RHEED intensity oscillations on a GaAs(001) substrate with the aid of a kSA-400 RHEED analysis system. The growth temperature was measured using an Ircon pyrometer.

The epi-ready semi-insulating GaAs(110) substrates misoriented towards (111)A by 2° (American Xtal Technology) were indium bonded to molybdenum disks and introduced into the MBE system without any ex-situ preparation. After initial outgassing at 350 °C for 1 h, the substrates were transferred to the growth chamber where the native oxide layer was removed at 620 °C under an As_4 flux of 2.5×10^{15} molecules cm^{-2} s^{-1}. After desorption of the native oxide, the initial broad-ringed RHEED pattern indicative of an amorphous surface layer was replaced by a clear 1×1 pattern. Subsequently, GaAs layers whose thickness ranged from 10 to 100nm (50, 100, 150, 200 and 500 ML in the [110] direction) were grown by MBE at a substrate temperature of 430 °C with a As/Ga flux ratio of 30 using a constant Ga flux of 8.75×10^{13} molecules cm^{-2} s^{-1}. Under these conditions, the GaAs growth rate on the (110) surface was 0.2 ML s^{-1}. During and after growth of the GaAs layers, the RHEED pattern displayed very sharp streaks in both [001] and $[1\bar{1}0]$ azimuths.

The surface morphology of the GaAs epitaxial layers was examined by atomic force microscopy (AFM) using a commercial instrument (Nanotec Electrónica S.L.) working in the constant force contact mode. Etched single-crystalline silicon nitride tips with a nominal end radius of 20 nm and a spring constant of 0.76 N/m were used with scan rates of 0.86 lines per second. Data were taken at 512 points per scanline. The mean values of the terrace width and the step height for each GaAs layer thickness were calculated from a set of over 20 cross-section profiles taken along the [001] direction from 3 μm × 3 μm AFM images of the GaAs(110) surface.

3. Results and discussion

Examination of the GaAs (110) surface by AFM immediately after desorption of the native oxide revealed the presence of 20 nm-deep pits of 250 nm mean diameter covering up to 20% of the surface area. The measured r.m.s. roughness was 4.23 nm. Fig. 1(a–e) shows a series of AFM images (3 μm × 3 μm) with their corresponding cross-section profiles along the [001] direction illustrating the GaAs(110) surface morphology evolution upon deposition of 50, 100, 150, 200 and 500 ML of GaAs at 430 °C with an As/Ga flux ratio of 30. The mean values of the terrace width (W) and the step height (H) are indicated in the cross-section profile graphs. Also shown in this figure is the 2D Fast Fourier Transform (FFT) obtained from each AFM image using a logarithmic normalization of the surface topography.

After deposition of the first 50 ML of GaAs there are some areas on the surface that have not been completely filled with GaAs material, which are related to pits on the original surface. However, at this early stage of growth bunched step structures with rather straight edges along $[1\bar{1}0]$ have already been formed with an average height of 5.5 nm. The terraces are bound by staircase step structures along the $[1\bar{1}2]$ directions, each having a height of 1–2 nm, as depicted in Fig. 1(a). The observed morphology suggests that adatoms diffuse more easily across $[1\bar{1}2]$-type step edges than across $[1\bar{1}0]$-type ones. Moreover, the incorporation of adatoms into $[1\bar{1}2]$-type step edges appear to occur at a faster rate than into $[1\bar{1}0]$-type steps causing the terraces to grow and merge laterally in the $[1\bar{1}0]$ direction with further growth (Fig. 1(b–e)). Thus, after deposition of 100 ML of GaAs, only a few staircase structures of $[1\bar{1}2]$-type steps are observed. With further growth, the average terrace width increases and the $[1\bar{1}0]$-type step edges show some waviness formed by the Bales–Zangwill instability [3]. Simultaneously, longer terraces along $[1\bar{1}0]$ are observed as the step bunching instability continues to develop.

Using a logarithmic normalization of the 2D FFT of the surface topography a symmetric pattern is obtained that evidences a certain periodicity between bunched step edges and in the step train inside each terrace. Up to 150 ML of GaAs the patterns are quite similar, but a significant change is observed when the ratio between the step heights and terrace widths becomes linear, a fact that occurs after 200 ML of GaAs have been deposited. A *transition stage* therefore takes place after deposition of approximately 150 ML of GaAs, according to the 2D FFT patterns shown in Fig. 1.

Analysis of the AFM data shown in Figs. 2 and 3 indicates that neither the terrace width distribution (TWD) nor the terrace height distribution (THD) fits to a Gaussian function in the initial stages of growth, but both evolve with time as the bunching instability develops. After deposition of 500 ML of GaAs the TWD exhibits a clear Gaussian behavior while the THD is very well fitted to a Lorentzian distribution. After deposition of 50 ML of GaAs the surface morphology shows a large dispersion in terrace height and width values. There is a clear predominance of terrace widths around 50 nm. The TWD can be fitted to the descending branch of a Lorentzian distribution or a bimodal distribution, which consists of a Lorentzian distribution centred on 50 nm and a Gaussian distribution centred on 170 nm. As GaAs is deposited, the terrace width mean value increases and the highest probability value shifts to larger values, fitting to a single-mode Lorentzian distribution (100–150 ML). Upon deposition of 200 ML of GaAs, a change in the distribution behavior is observed, fitting to a single-mode Gaussian distribution. As can be observed in