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Step Ordering during Fractional-Layer Superlattice Growth on GaAs(001) Vicinal Surfaces by Metalorganic Chemical Vapor Deposition

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Step ordering during fractional layer superlattice (FLS) growth is observed by transmission electron microscopy. A cross-sectional transmission electron micrograph of an $(AlAs)_{1/2}(GaAs)_{1/2}$ FLS shows that the steps of unequal spacings observed on a GaAs surface develop a sequence of uniformly spaced steps of single monolayer height during the growth. Comparison of the observed FLS structure with a numerical simulation based on the assumption of unequal incorporation probabilities of adatoms into up-steps and down-steps shows that a net flux into up-steps 2% larger than that into down-steps is enough to reproduce the FLS growth. We demonstrate that the FLS growth gives a unique opportunity to observe the step ordering process directly and determine the anisotropy of diffusion quantitatively.

1. Introduction

Step flow growth on a vicinal GaAs(001) surface has been used to fabricate quantum-wire-like structures or vertical superlattices such as tilted superlattices[1] or fractional layer superlattices (FLSs)[2]. New phenomena such as electron wave interference[3] and linear[4] and nonlinear[5] optical anisotropies have been observed in these structures. Step waviness and terrace width uniformity are the two most important factors that affect the quality of the structure grown on the vicinal surface. While step waviness is difficult to observe at the atomic scale without resorting to a sophisticated observation technique such as Scanning Tunneling Microscopy, terrace width uniformity can be observed with plan view or cross sectional-view Transmission Electron Microscopy (TEM). This presents an opportunity to compare theory and experiment and to obtain useful information about the step flow growth of GaAs and AlAs to improve the FLS quality.

As shown by Schwoebel,[6] a non-uniform step sequence in which terrace widths or step spacings are not equal develops an equi-distant step train as growth proceeds if atoms arriving on a terrace are incorporated more easily into the up-step than into the down-step bordering the terrace. Based on this model, we previously made a numerical calculation on the temporal evolution of a step sequence as growth proceeds.[7] Although we successfully demonstrated step ordering using a random distribution of step spacings as the initial condition through the numerical simulation, we were unable to make a reliable estimate of the asymmetry parameter that characterizes the asymmetry between up-steps and down-steps that causes step ordering because the TEM image of the FLS structure was blurred near the FLS/GaAs-buffer interface.

An improved MOCVD process using an extremely pure environment during growth and closely regulated flow control allowed us to observe how bunched steps near the FLS/GaAs-buffer develop a sequence of uniformly spaced steps of single monolayer height during $(AlAs)_{1/2}(GaAs)_{1/2}$FLS growth. From this observation, we quantitatively determine using the anisotropic diffusion model[7] the asymmetry in the incorporation of column III atoms diffusing on a terrace into the up-step and the down-step bordering the terrace.

2. Experimental

The FLSs were grown on a (001) GaAs substrate 2° tilted to the [T10] direction by MOCVD under higt AsH_3 and low triethylgallium (TEGa) or triethylaluminium (TEAl) partial pressure at 600°C. The growth was performed with a particle-free MOCVD system similar to the one described in Ref. 8. Additional improvements were a reduced He leak rate of lower than 10^{-10} atm cc/s and a lower dew point of H_2 carrier gas of -110°C. Otherwise, growth conditions were the same as before.[8] TEM observation for $(AlAs)_{1/2}(GaAs)_{1/2}$ FLS was performed from the [110] direction with a 300-keV incident electron beam.

3. Results and discussion

Figure 1 shows a [110] cross-sectional TEM view of

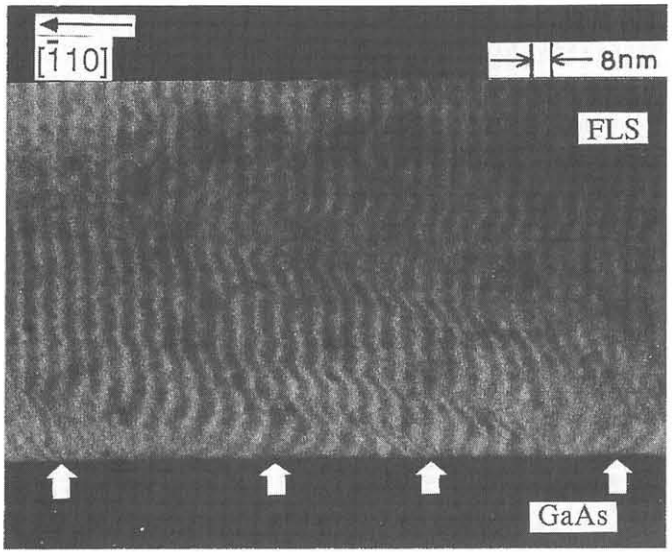

Figure 1. Cross-sectional TEM image of an $(AlAs)_{1/2}(GaAs)_{1/2}$ fractional layer superlattice grown on a GaAs (001) vicinal surface with steps of multiple monolayer height. White arrows indicate where multiple steps exist on the GaAs surface.

part of a 900-monolayer (250 nm) thick FLS grown at 600°C on a GaAs vicinal surface misoriented from (001) toward [T10] by 2°. Terraces are higher on the right. The FLS was grown on a 60-nm thick GaAs buffer layer, which appears as a dark portion lying under the FLS. The lighter region in the photograph corresponds to the AlAs-rich part and the darker region to the GaAs-rich part. The figure shows that a highly uniform FLS with a period of 8 nm and with high contrast between AlAs and GaAs develops as the growth proceeds on the surface of the GaAs buffer layer. Close examination of the FLS/GaAs-buffer layer interface reveals that the GaAs buffer layer shows a wavy surface with an amplitude of a few monolayers in height. Higher positions of the surface are indicated by white arrows in the figure. This indicates that the GaAs surface tends to develop a wavy or bunched surface with steps of multiple monolayer height spaced at 60-70 nm.[9] The FLS structure in the close vicinity of where the step bunching occurs on the GaAs buffer is tilted with respect to the (001) surface normal and its tilt angle varies depending on the position on the surface. Similar tilting and lateral distribution have been observed by Chalmers et al.[10] for (Al, Ga)Sb tilted superlattices. This is a natural consequence of bunched steps becoming spaced out during growth as demonstrated by numerical simulation in Ref. 7. Further examination shows that the step spacing becomes uniform after about 70 nm of FLS growth.

Qualitative agreement between the FLS structure observed near the FLS/GaAs-buffer interface and the simulated pattern shown in Ref. 7 prompted us to investigate the validity of the model discussed in Ref. 7 in a more quantitative manner. We chose a portion of the FLS structure that shows the characteristic pattern in which bunched steps develop more uniform step spacings and searched for the best-fit parameter that describes the anisotropy of incorporation of adatoms into steps.

The model assumes that the step flow velocity of a certain step V_1 is given by

$$V_1 = J_0\{ [(1+\eta)/2]\, T_1 + [(1-\eta)/2]\, T_0\},$$

where J_0 is the net flux onto the surface, η the anisotropy parameter representing the diffusion anisotropy of column III atoms near the step. T_0 and T_1 are widths of terraces on both sides of the step with terrace T_0 being one-monolayer higher than terrace T_1. If we know the terrace width distribution at a certain time t_0, we can use this equation to obtain a new step spacing distribution at time t_0+dt where dt is a small increment of time. For $\eta=0$, diffusion is isotropic near the step and the fluxes from terraces T_1 and T_0 both contribute equally to the step velocity, V_1; for $\eta=1$, V_1 is determined solely by the flux from T_1. The simulation shows that step spacing becomes uniform only when $\eta > 0$.

Figure 2 compares an enlarged TEM cross-sectional view and the simulated cross section. We used the step spacing distribution near the FLS/buffer interface determined from the TEM picture as the initial condition of the calculation. As noted above, the FLS is tilted with respect to the (001) surface normal near the GaAs buffer layer and the tilt angle varies as a function of position on the buffer surface. As the FLS growth proceeds, the FLS tilt angle from the surface normal decreases to zero and the tilt angle variation in the [T10] direction also diminishes, resulting in the formation of

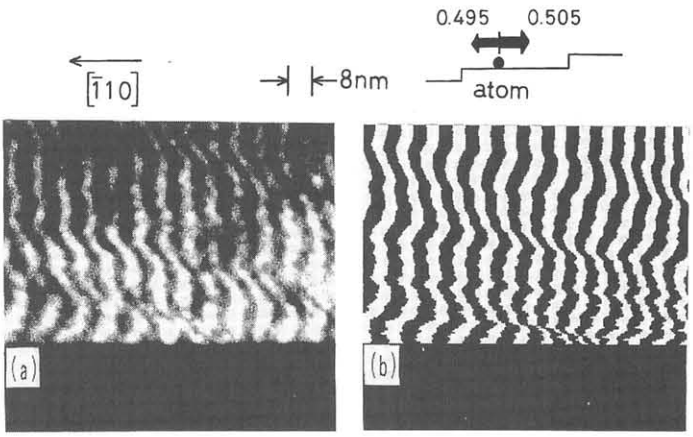

Figure 2. Comparison of TEM-observed step ordering and simulation result. (a) cross-sectional TEM image of an FLS, (b) simulated cross-section calculated using the anisotropic diffusion model for $\eta=0.01$.

an uniform FLS. This is the evidence of the step ordering process during the FLS growth. The result calculated with η as small as 0.01, which is shown in Fig. 2 (b), gives a good reproduction of the step ordering process near the multiple height steps on the buffer layer surface observed in the TEM picture. η equal to 0.01 means that the flux into the up-step is 2% larger than that into the down-step.

For η=0.03, the step ordering proceeds more rapidly than the observed results. Although the tilt angle of the FLS and its lateral distribution in the vicinity of the GaAs buffer layer surface, where the FLS thickness is less than 15 nm, are almost the same for η = 0.03 as for η =0.01, the FLS tilt angle around the center of the picture (about 30 nm from the interface) is 2/3 of that for η = 0.01, which is too small to reproduce the observed results. Furthermore, at 60 nm from the buffer layer surface, the FLS becomes nearly perpendicular to the surface in contradiction with the observed result. Thus η = 0.01 reproduces the TEM result better than η = 0.03.

Next we will consider whether it is valid or not to use a single time-independent asymmetry parameter η to simulate the FLS growth. Obviously, as the FLS growth consists of two alternate growth of GaAs and AlAs, we should in principle use at least two η's, one for the GaAs growth and one for the AlAs growth. However, it is necessary to determine the lateral position of the GaAs/AlAs interface at the atomic scale to obtain η's for GaAs and for AlAs independently. The present TEM picture of the FLS is not clear enough to determine the position precisely, so we can only get the average value of the two η's.

Next we discuss a possible mechanism of the step ordering during the FLS growth. We postulate that co-existence of Ga and Al is essential for step ordering because neither GaAs nor AlAs shows step ordering by itself. As observed by Kasu and Fukui[9] by Atomic Force Microscopy, GaAs steps tend to bunch as growth proceeds in MOCVD growth conditions similar to the ones used in our experiments. This is confirmed by the TEM observation shown in Fig. 1 as discussed above. We reproducibly observed step bunching on the GaAs surface. This indicates that η is negative for GaAs growth.

For AlAs growth, we reproduced the phenomenon observed in a previous experiment,[11] in which AlAs surface follows the ups and downs that exist on the substrate surface with grown thickness being the same everywhere. Although AsH_3 pressure was much higher in the present experiment than in the previous one, AlAs growth behavior was still essentially the same. These observations suggest that AlAs grows via two-dimensional nucleation. Therefore, the step spacings are independent of time and so is the surface profile. Al is considered to be more mobile on the GaAs part of an FLS surface than on the AlAs part, which helps Al adatoms to segregate on the AlAs rich region to form an FLS structure. This will not contribute to step ordering or step bunching.

Based on the above observations, a possible mechanism for steps to order is that η becomes positive for Ga adatoms migrating on AlAs, while Ga adatoms on GaAs have a negative η. The difference in the behavior of Ga adatoms on GaAs and AlAs might originate from the difference in the strength of the interaction between adatoms and an underlying layer or the difference in step waviness.

4. Conclusions

In conclusion we found by TEM that the steps of unequal spacings observed on a GaAs surface develop a sequence of uniformly spaced steps of single monolayer height during the $(AlAs)_{1/2}(GaAs)_{1/2}$ FLS growth. Comparison of the observed FLS structure with a numerical simulation based on the assumption of anisotropic incorporation of adatoms into up-steps and down-steps gave a quantitative estimate of the anisotropy, i.e., the flux into the up-step is 2% larger than that into the down-step. FLS growth presents a unique opportunity to observe the step ordering process directly and determine the anisotropy of diffusion quantitatively.

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