Surface Evolution during Molecular-Beam Epitaxy Deposition of GaAs

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Scanning tunneling microscopy studies have been performed on GaAs homoepitaxial films grown by molecular-beam epitaxy. Images show that in the earliest stages of deposition the morphology oscillates between one with two-dimensional islands and flat terraces. As growth proceeds there is a gradual coarsening of the surface features. Comparison with reflection high-energy electron diffraction (RHEED) leads us to propose that there is a direct correspondence between the surface step density and the RHEED specular intensity. As such, we associate the decay of the RHEED oscillation amplitude with a reduction in the temporal variation of the step density rather than the buildup of interface width.

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Since its introduction about 30 years ago, the molecular-beam epitaxy (MBE) growth technique has been utilized by both device engineers and scientists to great advantage [1]. It affords monatomic layer thickness control over films growing from the vapor phase at relatively low temperatures under supersaturation conditions. The formation of new metastable phases has been realized. Additionally, the highly nonequilibrium situation which exists in MBE has enabled the creation of multilayered structures in which individual layers maintain their chemical integrity and form compositionally abrupt interfaces with one another. This has led to the fabrication of heterostructures and superlattices, which, in the case of semiconductor systems, possess novel electrical and optical properties. For optimum performance of devices based on these artificial materials, the morphological sharpness of the interfaces is of primary importance. Roughness leads to increased carrier scattering in active regions, which lowers electron and hole mobilities. It also causes variations in the quantum well width which broaden exciton linewidths. As interface smoothness is ultimately determined by surface morphology, it is technologically important to understand the mechanisms controlling morphology.

From an atomistic perspective, the surface morphology of a growing film is controlled by surface kinetic processes: dissociation, chemisorption, desorption, diffusion, step-edge attachment, and 2D nucleation are the most basic ones; necessarily, some of these processes must occur before an atom or molecule from the vapor is incorporated at the growth front. Under the normal supersaturation conditions which exist for MBE growth, surface kinetics is significantly limited, causing growth to occur far from a 2D quasiequilibrium state. Consequently, even in the simplest of material systems, homoepitaxy, an understanding of the dynamical evolution of the surface morphology is challenging.

Reflection high-energy electron diffraction (RHEED) has become the standard tool for characterization of films during MBE growth [2]. The geometry of a grazing incidence angle for electrons results in surface sensitivity and enables the growth front morphology to be monitored during deposition. In addition to a straightforward determination of the surface symmetry from the diffraction pattern, indirect statistical information of surface disorder is obtained from modulations in intensity along particular diffraction features. However, since electrons interact strongly with matter, dynamical scattering effects must be included in a complete analysis aimed at relating modulations or absolute intensities to surface disorder. Only a limited amount of theoretical work has been undertaken on this subject, making it difficult to accurately determine the surface morphology from RHEED. Nevertheless, RHEED measurements during homoepitaxy have revealed a striking feature related to the growth process. Under many conditions, upon initiation or reinitiation of growth, there are transient temporal oscillations in the intensity of the diffraction features. The period of the oscillations corresponds to the time to deposit a monolayer. Although the behavior of the oscillations depends on growth and diffraction conditions, the effect is generally considered to be a manifestation of a 2D layer-by-layer growth [3].

As a result of the RHEED observations, models addressing the epitaxial growth of GaAs(001) naturally attempt to compare with experiment by determining an oscillating quantity. Although considerable effort has been made in this regard, the question "What is oscillating?" still remains. This issue is broader than simply determining what RHEED measures. RHEED oscillations reveal a general pattern or characteristic in the growth behavior. Consequently, correlating the diffraction signature with the surface morphology quite generally advances an understanding of epitaxial growth. Efforts aimed at modeling nonequilibrium film growth phenomena are particularly hindered by the lack of a real space picture of the growth front as it evolves.

In this Letter we present scanning tunneling microscopy (MBE) images of GaAs(001) surfaces as they appeared during deposition. The goal of these experimental studies is to obtain a real-space picture of the evolution of the surface during growth and investigate its relationship to RHEED oscillations. The experiment consists of initiating growth from a recovered surface and then ter-

minating deposition at a specific point during the growth process. This procedure is then repeated for various termination points. Because the samples are removed from the STM for regrowth, no direct comparison can be made between any specific feature in the progression of the images. We have imaged large areas at multiple sites on multiple samples; the images shown are representative of the surface. There appears to be very little inhomogeneity in the samples.

Deposition was performed in a standard ultrahigh vacuum system, base pressure 7×10^{-11} T. Effusion cells were used to produce both the Ga and As_4 fluxes. Commercial GaAs(001) substrates were first chemically cleaned, then loaded in the vacuum system where the oxide was removed at 580 °C under an As_4 flux. To smooth the substrate a 300-nm-thick buffer layer was grown. The substrate temperature during deposition was 555 °C. The As to Ga pressure ratio was 15 and the deposition rate was 0.15 monolayer/sec. The sample miscut as determined by STM was approximately 0.15°. The direction and magnitude of the local vicinality was found to vary appreciably. The incident angle of the RHEED beam was approximately 0.9° and corresponded closely to the "in-phase" Bragg condition. This diffraction condition was chosen to yield a reduction in specularly scattered electron intensity with the initiation of growth. The azimuthal angle corresponded to a beam direction parallel to [110].

A central technical point of the experiment was the ability to quench the surface morphology as it appeared during growth. To accomplish this a resistively heated low thermal mass sample was used. The sample temperature could be reduced from that during growth (555 °C) to below 450 °C in 1.5 sec. A liquid-nitrogen-cooled baffle with a cooled shutter shrouds the sample [4]. The quench procedure proceeded as follows: close the Ga shutter, reduce the sample temperature to between 400–450 °C, close the liquid-nitrogen-cooled shutter (total elapsed time 2 sec), turn off the Ga and As sources, allow the background As pressure to drop into the low 10^{-9}-T range (~30 min), turn off the sample heating, and transfer into the STM chamber. The RHEED intensity was recorded up to the time the transfer occurs. We found that the RHEED intensity is quite steady during this period. Holding the sample at 400 °C appeared to eliminate physisorption of As while not allowing significant step edge motion [5].

Figure 1 shows the RHEED curves taken during a quench at the fourth intensity maximum and minimum. The constant RHEED intensity after quenching indicates that the surface evolution has been halted. Figure 2(a) shows an STM image of a 200-nm×200-nm area of a recovered surface. The terrace size is large and the step edges smooth. This is the GaAs surface as it appears be-

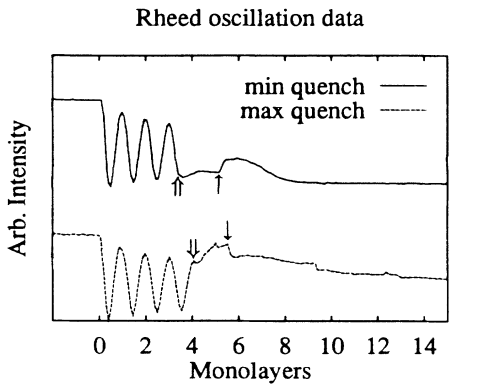

FIG. 1. RHEED specular intensity oscillations for GaAs on GaAs(001). The incident angle was 0.9° and the azimuth was along [110]. The ⇓ labels the point at which growth was terminated for each sample and the ↓ indicates an experimental artifact due to the quenching procedure. STM data presented in Figs. 2(b) and 2(c) were acquired from these samples.

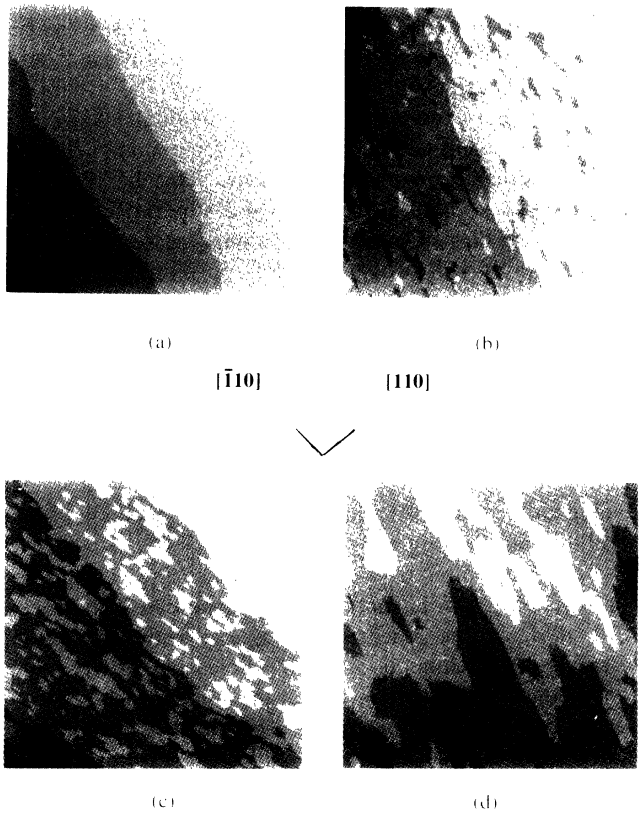

FIG. 2. (a) STM image of a GaAs(001) buffer layer. The scan range is 200 nm×200 nm. The tunneling voltage (V_t) was +2.8 V, applied to the sample, and the tunneling current (I_t) was 80 pA. (b) STM image of GaAs(001) surface after termination of growth at a RHEED maximum. (c) STM image of GaAs(001) surface after termination of growth at a RHEED minimum. (d) STM image of GaAs(001) after 60 monolayers have been deposited. Note that the local direction of the vicinality in (d) differs from that shown in the previous images. (b) and (c) correspond to the traces presented in Fig. 1.

fore growth. Figures 2(b) and 2(c) are images of the surface as it appeared at the intensity maximum, and the intensity minimum shown in Fig. 1.

There is clearly a morphological change in the sample surface profile as it evolves from a RHEED maximum to a RHEED minimum. The surface quenched at an intensity maximum shows few islands on terraces and an approximately equal number of monolayer deep holes. In contrast, the surface quenched at a RHEED minimum shows many two-dimensional islands on the terraces. There is a much lower density of monolayer deep holes. The terrace edges for both samples are relatively smooth.

In order to correlate the morphological evolution we observe with the time variation of the RHEED specular intensity one must consider various proposed models for the interpretation of the diffraction process. One simple approach uses a kinematical approximation to determine the interaction of the scattered electrons with the surface [6]. In this picture the measured intensity is due to the interference of the electrons scattered from different terraces on the surface. At the correct incident angle (off-Bragg condition) this leads to an oscillation of the specularly reflected intensity due to a changing terrace occupation during deposition. As each growing layer proceeds from zero coverage through half filling and finally to a complete layer the specular intensity cycles through one period. A second, largely phenomenological, model which attempts to incorporate diffuse scattering has been proposed to explain RHEED oscillations. In this model the relevant quantity is not the terrace occupation but the step edge length per unit area, termed step density [7]. Steps provide a mechanism for diffuse scattering of the electron beam. With an increase in the step density the specularly reflected intensity would decrease. As in the previous model, if the surface morphology cycles from islanded to flat then the RHEED intensity would vary accordingly. Monte Carlo growth simulations have shown an excellent correspondence between the step density and experimental RHEED data taken on vicinal surfaces [8]. The third explanation for RHEED oscillations during GaAs growth involves the dissociation dynamics of As_2 of the surface [9]. By postulating a configuration-dependent reaction rate it is found that the As_2 dissociation displayed an oscillatory behavior with the correct monolayer period. This model does not require that there be any differences in morphological step distribution during growth to account for the intensity oscillations.

There has been criticism of these models due to the incomplete treatment of dynamical scattering. It has been experimentally demonstrated that the behavior of the scattered electrons is a complicated function of both azimuthal and polar angles [10]. These data cannot be explained within a kinematical framework. In recent years some progress has been made towards a fully dynamical approach [11]. However, the surfaces so treated have been highly idealized and the connection with real systems remains tenuous.

Our STM data can be interpreted within the context of the step density model. The diffraction conditions correspond to the "in phase" Bragg condition where electrons reflected from adjacent terraces constructively interfere. The specular intensity varies not because of interference but, because of diffuse scattering from step edges [12]. Table I presents the surface step density, and rms roughness for various film thicknesses. There is a correspondence between films with higher step density and lower RHEED specular intensity. On closer inspection of the data it is natural to ask if the scattering from islands and holes should be weighted equally. Geometrically, the scattering from a small hole might be quite different than that from an island or terrace edge. We have calculated the specular scattering from holes and islands for the Bragg condition within a modified Born approximation [13]. It is found that holes less than 5 nm in diameter contribute far less to the diffuse scattering than do islands and terrace edges. As a first approximation, if the step density contribution from small holes is subtracted from the total then the agreement between this modified step density and RHEED intensity is quite strong, see Table I. Although the exact functional dependence of the intensity on step density is probably complicated, the physical mechanism of diffuse scattering from steps is clearly indicated.

To further investigate the growth process, we have also examined the sample surface after the decay of RHEED oscillations. Figure 2(d) shows the surface after 60 monolayers have been deposited and the RHEED oscillation amplitude has decayed to less than 5% of its original value. The sample is quenched in the same manner as before. The typical feature size has increased and in contrast to the earlier data the two-dimensional islands and terrace edges are now quite ramified. As shown in Table I the rms roughness of the surface has not changed from that measured for the films where intensity oscillations were observed. This points to a central feature of the data, the decay of the oscillation amplitude has occurred

TABLE I. Step densities and interface widths for various layer thicknesses. The modified step density has the contribution from small (< 5 nm diameter) holes eliminated. The step density is defined as the step edge length per unit area and the rms roughness is defined as $[N^{-1}\sum_i (h_i - \bar{h})^2]^{1/2}$ where h_i is the height and the sum is over a 200-nm × 200-nm area.

	Film thickness (GaAs bilayers)				
	0	0.25	3.5	4.0	6.0
Step density (10^{-2} nm^{-1})	2.0	9.3	13.8	8.9	7.6
Modified step density (10^{-2} nm^{-1})	2.0	9.0	13.1	5.6	7.3
rms roughness (nm)	0.10	0.16	0.16	0.14	0.17

without an increase in the interface roughness. The surface has achieved a steady-state configuration, through a balance of island nucleation and step-flow, which evolves with a constant step density. For these diffraction conditions increased surface roughness is not the cause for the RHEED oscillation decay. Recent Monte Carlo simulations agree with these data [14].

In summary, we have studied the evolution of the GaAs surface during MBE growth. Starting from a recovered substrate, upon the initiation of growth the surface progresses through a transient regime, where cyclic changes in the step density are found, to a steady state. There is a clear connection between the surface morphology and RHEED oscillations. With in-phase diffraction conditions, chosen to eliminate the kinematic contribution, the RHEED intensity oscillations are shown to correlate well with the surface step density. A most striking result is that the decay of the RHEED oscillations is not due to an increase in surface width, but the evolution of the surface to a steady state with constant step density.

As a possible refinement of the step density model one should consider the effects of anisotropy. The STM images of the GaAs surface show islands and monolayer holes greatly elongated along $[\bar{1}10]$ direction. Perhaps the relevant step edge length is the component perpendicular to the incident electron beam. The step density analysis is certainly an oversimplification to a complete multiple scattering theory. What is the nature of the scattering from the adatoms? The notion that scattering from small holes may be different from islands points to the need for detailed calculations which include dynamical scattering from different morphologies. In spite of these questions, for the diffraction conditions used during this study it appears that the phenomenological step density model shows substantial agreement with experiment.

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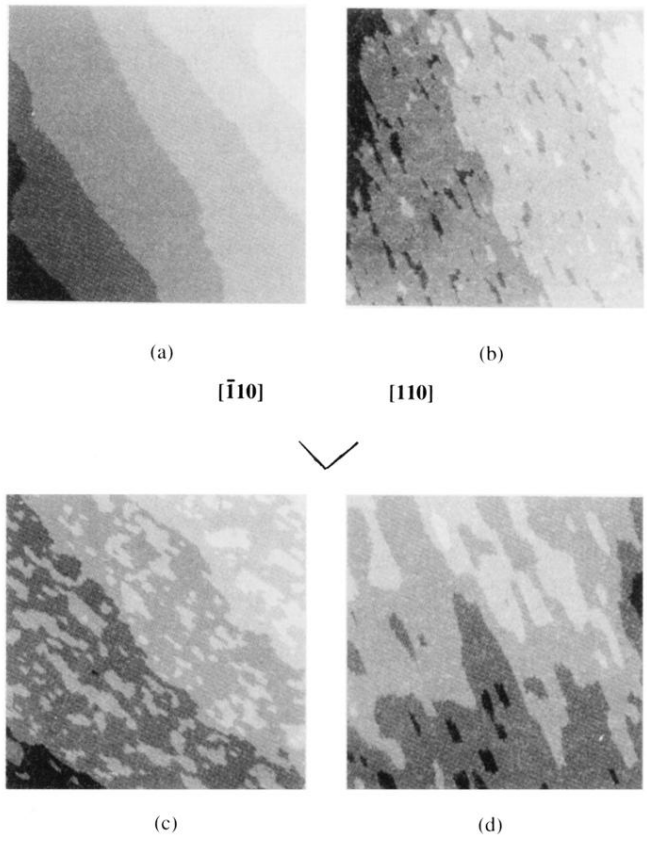

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