

## Influence of surface step density on reflection high-energy-electron diffraction specular intensity during epitaxial growth

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Growth by molecular-beam epitaxy is modeled by Monte Carlo simulations of a two-step process that includes deposition and migration. As a marked departure from previous studies, we monitor the evolution of the growth by calculating the surface step density, which we then compare with the specular intensity in reflection high-energy-electron diffraction measurements. The strong correspondence between the two quantities facilitates considerable insight into the microscopic origins of the influence of various externally controlled parameters upon the high-energy-electron diffraction profile.

Reflection high-energy-electron diffraction (RHEED) has emerged as the routinely used experimental probe for *in situ* analysis of molecular-beam epitaxy (MBE). The observation of periodic oscillations in the spectral RHEED beam intensity during growth (Fig. 1) offers a reliable measure of the growth rate and has provided the most direct evidence for the layer-by-layer growth mode.<sup>1-3</sup> Additionally, RHEED measurements have been used to explore surface diffusion,<sup>4</sup> dopant incorporation,<sup>5</sup> and growth front relaxation upon termination of the beam flux.<sup>6</sup> Previous theoretical studies of RHEED from growing surfaces have either utilized the kinematic approximation to diffraction on simulated growth fronts<sup>7</sup> or have employed a full dynamical treatment using considerably simplified atomic configurations.<sup>8,9</sup> Though these studies stress the dependence of the specular RHEED intensity on the step distribution of the surface, the absence of full dynamical calculations for realistic growth fronts has often resulted in conflicting interpretations of RHEED data.<sup>8-10</sup>

We have taken a somewhat different approach to the investigation of RHEED during growth by MBE with the application of a Monte Carlo simulation of epitaxial growth and continuous monitoring of the surface step density of the sample. Our previous work has shown that the evolution of surface step densities shows a remarkable

correspondence to measured RHEED data for growth on both flat<sup>11</sup> and stepped<sup>12</sup> surfaces, including the effects of relaxation.<sup>6</sup> Here, we examine this correspondence more closely and conclude that the efficacy of the step density as a simulation of RHEED measurements arises from the effective inclusion of multiple-scattering corrections to the kinematic theory, which is most apparent at half-layer completion. The development of an approach that simulates not only *growth* by MBE, but also the *monitoring* of the growth by RHEED, opens the way to many applications that would otherwise have been difficult without full dynamical RHEED calculations on disordered surfaces.

Our studies have been carried out on a model that consists of two basic steps: (1) random deposition of atoms onto a finite substrate with periodic boundary conditions and (2) the migration of surface adatoms. Evaporation is neglected as under typical growth conditions; the desorption of adatoms from the surface has been shown to be negligible. Atoms are generated at random points on the lattice, with an interarrival time  $\delta t = (FA)^{-1}$ , where  $F$  is the flux of incident species and  $A$  the area of the substrate. Diffusion is modeled as a random hopping process with individual atoms having a nearest-neighbor intersite hopping rate given by the Arrhenius form  $D(T_s) = (2k_B T_s/h) \exp(-E_D/k_B T_s)$ , where  $T_s$  is the substrate temperature and  $E_D$  the binding energy of the atom under consideration. The diffusion barrier  $E_D$  is comprised of two terms:  $E_D = E_s + nE_b$ , with  $E_s$  being the barrier to diffusion of a free atom,  $E_b$  the energy of each bond formed with a nearest neighbor along the surface, and  $n = 0, \dots, 4$  the number of such bonds. In the interests of economizing on computer resources we make an approximation to diffusion that allows adatoms with only one or no nearest neighbors the opportunity to migrate. Our studies have shown that this has a negligible effect on the evolution of the surface during growth, though in the post-growth relaxation phase the approximation is no longer valid.

The simulations we present below were carried out on a  $30 \times 30$  square lattice with periodic boundary conditions, a lattice constant of  $5.5 \text{ \AA}$  and a flux  $F = 1 \times 10^{18} \text{ m}^{-2} \text{ s}^{-1}$ , yielding a growth rate of  $0.3$  monolayers/sec. Tests of convergence with increasing lattice size revealed that the

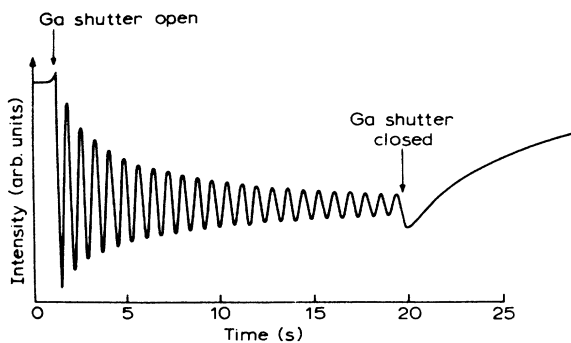


FIG. 1. Typical measured RHEED intensity evolution, in this case for the growth of GaAs (after Ref. 14).

characteristic features of the model are well represented on a  $30 \times 30$  lattice. The values of the energy parameters were chosen as  $E_s = 1.3$  eV and  $E_b = 0.5$  eV.

In Fig. 2 we show typical growth fronts at four substrate temperatures,  $T_s = 500, 610, 630,$  and  $650$  K. Each simulation has been run for the same real time, so that the same number of atoms has been deposited, in this case enough for twenty complete layers. We note two important features: (1) the effects of enhanced surface diffusion are apparent by the increased size and smaller number of growing clusters at higher temperatures and (2) the better growth quality at higher temperatures, in the sense of there being a smaller fraction of incomplete layers. In particular, the growth pattern is clearly three dimensional at  $500$  K, but essentially two dimensional at  $650$  K.

To make direct comparisons between our simulations and RHEED data, we show in Fig. 3 the calculated step densities as a function of time at the four temperatures shown in Fig. 2. Since a larger step density implies a lower specular intensity, we have inverted the ordinate of the graphs in Fig. 3 to coincide with the usual RHEED intensity plots. We see that growth at  $500$  K shows no sign of the layer-by-layer mode and decays rapidly to a steady-state (nonequilibrium) roughness. The growth at  $610$  K is qualitatively different from that at  $500$  K, as

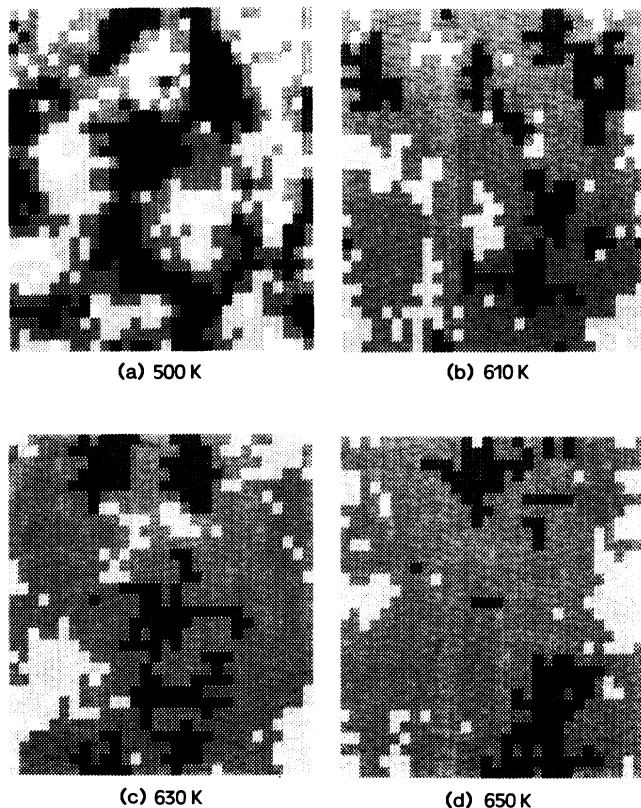


FIG. 2. Growth configurations for individual simulations at the indicated substrate temperatures after the deposition of enough atoms for 20 complete layers. The  $n$ th partial layer is indicated by shading which darkens with increasing  $n$ .

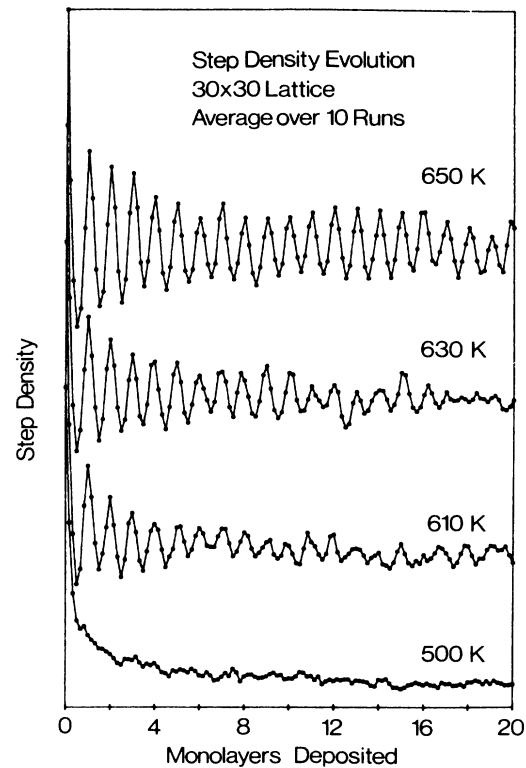


FIG. 3. The step density plotted as a function of time for the indicated substrate temperatures. Note the inverted ordinate axis.

would be expected from a comparison of Figs. 2(a) and 2(b). The rapidly decaying envelope and irregularity of the oscillations indicate that the layer-by-layer growth mode is unstable, although as can be seen in Fig. 2(b), even after deposition of 20 layers, the surface is still flat to within an accuracy of one monolayer, in contrast with Fig. 2(a) which clearly is three dimensional.

The evolutions of the step density at  $630$  and  $650$  K show much more regularity than at lower temperatures, displaying a decaying envelope in the step density at both layer completion and at half-layer completion. The step density shows a marked correspondence with measured RHEED evolutions (Fig. 1), in contrast to calculations based upon kinematic theory, which exhibit a decay in the intensity at the layer completion points but *not* at the half-layer completion points. Taking into account the finite solid angle of the aperture yields no better agreement for this aspect of RHEED data at the kinematic level.<sup>13</sup> This failure of the kinematic theory may be readily explained by the fact that at half-layer completion the surface shows the greatest step density, so that multiple scattering between steps would be most likely, rendering single-scattering theory inadequate. Furthermore, in the kinematic approximation, the specular RHEED intensity is sensitive only to coverage, not to specific configurations, which again is quite a serious drawback at half-layer completion. On the other hand, the step density contains more

configurational information than surface coverage and thus in effect provides some corrections to the kinematic theory due to multiple scattering.

Having established the correspondence between the step density evolution calculated in our model and RHEED measurements, we turn our attention to the physical origin of the decaying envelope, which has often been regarded as indicative of an eventual transition to a three-dimensional growth mode. The final surfaces for  $T_s = 610$  and 630 K are clearly two dimensional to within one-monolayer accuracy. The eventual loss of the oscillations may be explained by the observation that regions separated by distances much greater than the surface diffusion length are statistically independent, a situation which can also result from absorbing discontinuities in the surface topography, such as step edges and immobile point defects.<sup>11</sup> Thus differences in the respective stages of growth can develop, though each region may be growing in a layer-by-layer fashion. These differences are manifested in the evolution toward a steady-state step density, resulting from an incoherent superposition of individual oscillatory step densities. Figure 4 shows five individual (statistically independent) step-density evolutions for  $T = 630$  K, before averaging to produce the result displayed in Fig. 3. Inspection of the individual step-density evolutions shows that different samples do in fact show phase differences in the step-density evolution at given times, leading to the loss of global resolution described above.

As a further demonstration of this observation we have simulated growth at 610 K on a  $90 \times 90$  sample, equivalent in area to nine  $30 \times 30$  samples and found that the behavior of the step density is essentially the same as that shown in Fig. 2, the oscillations becoming insignificant after deposition of  $\approx 6$  monolayers. The well-defined oscillations thus appear to be the result of the initial conditions, i.e., a perfectly flat surface, and the eventual decay of the oscillations implies only the development of a steady-state step density, and certainly *not* a transition to three-dimensional growth.

To summarize, we have found that the application of a simple schematic model of MBE growth combined with a calculation of the surface step density reproduces the principal features of measured RHEED intensity oscillations (which we would thereby expect to be of a universal nature). The kinematic approach to the calculation of

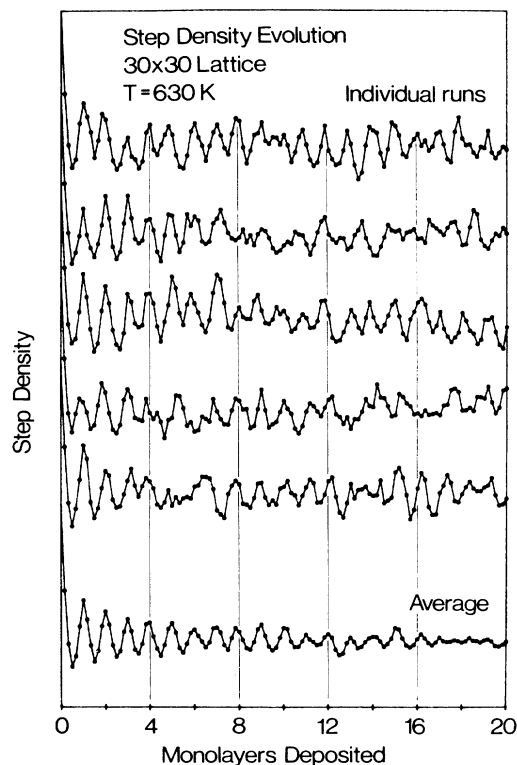


FIG. 4. Step-density evolutions for five individual samples (with no averaging) at  $T = 630$  K.

RHEED from growing surfaces is shown to be inadequate in the high-disorder regime while the step density shows very good agreement over the full range of growth stages. Furthermore, the simplicity of the model facilitates the study of epitaxial growth with realistic deposition rates for extended periods of time, the importance of which for the exploitation of MBE is evident.

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