Anisotropic kinetics and bilayer epitaxial growth of Si(001)

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The epitaxial growth kinetics of Si(001) are investigated by the application of a Monte Carlo simulation, with explicit incorporation of the anisotropic bonding induced by the (2×1) dimer reconstruction. The model produces a pseudobilayer growth mode on a single-domain substrate and resolves apparently contradictory observations of Si homoepitaxial growth reported earlier by different groups.

The integration of recently developed III-V optoelectronic semiconductor devices with Si microcircuitry has profound implications for the development of advanced telecommunications and even optical computers. This has prompted considerable interest in homoepitaxial and heteroepitaxial growth on Si substrates by molecularbeam epitaxy (MBE). In fact, Si MBE is intriguing in its own right, as reported work indicates behavior which not only conflicts with currently accepted models for growth by MBE developed for III-V compounds,^{1,2} but even at first sight appear to be contradictory. In this paper, we develop a model for Si MBE which suggests that the apparent discrepancies reported for epitaxial growth of Si(001) may be traced to the dimer reconstruction, which induces a local anisotropy in surface bonding and whose global structure is extremely sensitive to annealing. When combined with our earlier work, the results reported here provide a convincing qualitative picture of the growth kinetics of group-IV MBE.

The structure of vicinal Si(001) has been studied by a variety of techniques, from which reconstructions have been identified that comprise rows of surface dimers in ordered (2×1) and (1×2) configurations. Prolonged annealing on a vicinal substrate (misorientation angle $2^{\circ}-4^{\circ}$) at high temperatures yields a single-domain (2×1) reconstruction with terraces separated by biatomic steps.^{3,4} Otherwise, the surface shows both (2×1) and (1×2) domains, with antiphase boundaries formed at monatomic steps.

The following features of Si(001) homoepitaxy have been reported.

(i) On a single-domain substrate Sakamoto *et al.*³ measured both monolayer- and bilayer-period reflection high-energy electron-diffraction (RHEED) oscillations, depending on the azimuthal direction of the electron beam. This study also revealed the remarkable stability of the bilayer oscillations, with 2200 periods being observed.

(ii) Conversely, in a later study, Aarts, Gerits, and Larsen⁵ reported strongly decaying monatomic RHEED intensity oscillations, evident for approximately 20 periods, with no observable azimuthal phase dependence. The substrate initially exhibited both (1×2) and (2×1) domains. (iii) Aarts, Gerits, and Larsen⁵ also noted the absence of any recovery upon cessation of the molecular-beam flux at low temperatures (≈ 300 °C). This suggests the presence of a layer-by-layer growth mode, with essentially immobile adatoms.

Feature (iii) has been addressed in an earlier publication.⁶ Here we focus upon (i) and (ii).

The structural characteristics of Si(001) have important implications for homoepitaxial growth. The (2×1) reconstruction leads to enhanced bonding perpendicular to the dimerization, as shown by calculations of step energies.⁴ We show here that feature (i) is a natural consequence of anisotropic bonding in Si(001) and that the discrepancy between (i) and (ii) may be resolved by considering the effect of competing anisotropies in coexistent (1×2) and (2×1) domains.

We employ a Monte Carlo simulation of epitaxial growth based upon a solid-on-solid (SOS) model, wherein the substrate is described as a simple-cubic lattice with neither vacancies nor overhangs. The growth process comprises two steps: random deposition of incident atoms from the molecular beam, and the migration of surface adatoms. The long residence time of Si atoms, $\approx 10^7$ s⁷, enables us to neglect the effect of evaporation. Surface migration is modeled by an Arrhenius expression for the intersite hopping rate: k(E,T) $=k_0 \exp(-E/k_B T)$, where k_0 is the vibrational frequency of a surface adatom, k_B is Boltzmann's constant, T is the substrate temperature, and E is the local energy barrier to hopping. We introduce an anisotropy into this energy barrier with the following properties.

(1) On a given layer, in-plane nearest-neighbor bonding is enhanced perpendicular to the direction of the dimerbond axis. This accounts for the difference in step energies resulting from the formation of surface dimers.⁴

(2) The anisotropy due to dimer formation rotates through 90° with each successive monolayer.⁸

These effects are introduced into the simulation through the hopping rate. The bonding anisotropy (1) appears in the energy barrier to diffusion,

$$E = E_S + mE_{\parallel} + nE_{\perp} ,$$

where E_S is the substrate contribution, E_{\parallel} and E_{\perp} are

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nearest-neighbor contributions parallel and perpendicular to the direction of bond enhancement, and m and n are the associated numbers of nearest neighbors, respectively. We have set the surface energy barrier, $E_S = 1.3$ eV, com-parable to measured values of $1.1,^7 1.3,^9$ and 1.57 eV.¹⁰ and we have chosen $E_{\parallel} + E_{\perp} = 0.5$ eV, in accord with our previous studies for isotropic growth.^{2,11,12} The ratio $E_{\parallel}:E_{\perp}$ was determined by selecting values that best prolonged the layer-by-layer growth mode, a characteristic feature of Si(001) MBE.³ Application of this procedure led to the selection of a 10:1 ratio. Upon selecting an atom for diffusion, the direction is generated both randomly and isotropically. There is almost certainly an anisotropy in the direction of migration, though this has not yet been observed experimentally and we find the presence of such an effect to be negligible in comparison with that induced by the bonding. Thus, to avoid overburdening our model with indeterminate parameters, we ignore this effect; it may be easily included once experimental evidence is available.

In having adopted a simple-cubic model of Si(001) with rules (1) and (2) for diffusion and bond formation, we have effectively constructed a "mean-field" description of Si MBE. Structural fluctuations, such as local deviations from the (2×1) dimer configuration,¹³ could by-pass the anisotropy of Si(001) as formulated above. However, our results are correct on the average.

Comparison of simulation with experiment is achieved by continuous monitoring of the surface step density, a quantity previously shown to yield good qualitative agreement with specular RHEED measurements,^{2,6,11,12}

$$S(\phi) = \frac{1}{L} \sum_{i,j} \{ [1 - \delta(h_{i,j}, h_{i+1,j})] \cos \phi + [1 - \delta(h_{i,j}, h_{i+1,j})] \sin \phi \}$$

where $\delta(a,b)$ is the Kronecker δ function and L is the number of lattice sites; here we have imposed periodic boundary conditions. We have introduced an angular dependence ϕ into the step density in order to allow comparison with the azimuthal resolution of the RHEED measurements.^{3,14} In displaying the data we have inverted the step-density evolutions to allow ease of comparison with RHEED data, a large step density corresponding to a low RHEED intensity.

In Fig. 1 we show step-density evolutions calculated in the $\phi = 45^{\circ}$ and 90° azimuths for the growth of 20 monolayers (ML) at T=700 K and flux F=1 ML/s. The toppanel data [Fig. 1(a)] were obtained from a surface with four monoatomic steps, representative of coexistent (2×1) and (2×1) domains, while that in the lower panel [Fig. 1(b)] is from a surface with two biatomic steps, representative of a single-domain surface.

Addressing the single-domain evolutions first [Fig. 1(b)], we observe an azimuthal dependence in the stepdensity calculation, a monolayer period in the $\phi=45^{\circ}$ direction ($\langle 100 \rangle$ azimuth), but a bilayer mode in the $\phi=90^{\circ}$ direction ($\langle 110 \rangle$ azimuth). We also note the enhanced stability of the bilayer oscillations, prolonged oscillations for 80 monolayers being readily observable (Fig. 2). At a higher substrate temperature of T=750 K



FIG. 1. Step-density evolutions projected along the indicated azimuths for growth on (a) a two-domain surface and (b) a single-domain surface. (Note that the ordinate has been inverted.)



FIG. 2. Prolonged step-density oscillations on a single-domain substrate.



(a) 1.5 Monolayers Deposited



(b) 2.5 Monolayers Deposited

FIG. 3. 60×60 subsections of a single-domain surface, obtained during the simulation represented in Fig. 1(b). Increasing height is represented by decreasing darkness.

extended growth runs of over 1000 monolayers still exhibits oscillation in the $\phi = 90^{\circ}$ azimuth.

The surface morphology of the growing crystal is illustrated in Fig. 3 for 1.50 and 2.50 ML deposition at T=700 K. It is evident that the growing clusters are elongated and change orientation on successive monolayers. Recent scanning-tunneling-microscope (STM) studies of the Si(001) surface after growth show strikingly similar elongation and reorientation.^{15,16}

The origin of the bilayer oscillations may be deduced by considering the morphology of the surface. Figure

3(a) shows clusters elongated along the y axis, yielding a high step density in the $\phi = 0^{\circ}$ azimuth. A monolayer later, the clusters are elongated along the x axis and we observe a low step density in $\phi = 0^\circ$. Consequently, we observe bilayer oscillations with minima and maxima both corresponding to half-layer completion. The increased stability and amplitude of the bilayer oscillations over the monolayer oscillations may also be explained by invoking the anisotropy in surface island structure.^{15,16} When resolved onto a nearest-neighbor bond direction, the step density is no longer sensitive to layer completion, but merely to the density of islands elongated in a given direction. Thus, provided the surface maintains a preponderance of one domain over the other, oscillations will be observed. The elongation will also lead to an enhanced stability of the oscillations in all azimuths, with atoms easily diffusing across the shortest cluster radius, leading to a higher degree of layer completion prior to the initiation of the next monolayer.

We now turn our attention to the contradictory results of Aarts *et al.*,⁵ who observed strongly decaying monatomic oscillations in the RHEED specular intensity during Si MBE upon a $(2 \times 1)/(1 \times 2)$ (001) surface. Analogous with RHEED behavior, the step-density oscillations on a double-domain surface [Fig. 1(a)] exhibit a monolayer period, are strongly damped, and have no azimuthal phase dependence. Evidently, with both domains present we sample an average of the two orientations, thus always displaying the monolayer growth mode. Hence, unless the substrate is annealed at high temperature prior to growth, to obtain a single-domain surface³ observation of prolonged oscillations will not be possible.

In conclusion, we have demonstrated that explicit inclusion of the microscopic anisotropy of the Si(001) surface provides a model that accounts for experimental RHEED studies of Si MBE and, in addition, yields a detailed reproduction of the surface structure as observed by STM. When combined with our earlier work reproducing low-temperature growth modes⁶ and layercompletion dependences in the RHEED recovery profile¹⁷ during interrupted growth,¹⁴ the model provides a conceptually simple framework upon which more detailed investigations may be based. It is, however, an important observation that our schematic model contains sufficient structural and kinetic detail to reproduce a comprehensive catalogue of experimental phenomena. The development and application of models such as this is essential if the study of MBE is to proceed in a systematic fashion.

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