Step barrier for interlayer-diffusion in Fe/Fe(100) epitaxial growth

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The interlayer diffusion barrier for Fe/Fe(100) deposition is estimated by comparing the results of kinetic Monte Carlo simulations with experimental results in the first few monolayers of growth. We find that, in contrast to previous theoretical estimates for other systems, the step barrier for Fe/Fe(100) is small in comparison with the activation energy for diffusion on a flat terrace (0.454 eV). Our results resolve a long-standing controversy and provide quantitative support for the conjecture that the existence of mounds in this system is due to a finite positive step barrier.

Molecular-beam epitaxy (MBE) is an important technological process for the fabrication of nanostructures and high-purity crystals. Recent theoretical work on understanding MBE has focused on the roles of nucleation and diffusion in epitaxial growth. In particular, it is now recognized that one of the most important factors controlling the growth morphology and quality of epitaxially grown crystals is a barrier to diffusion of atoms between different layers. The existence of this barrier causes monomer diffusion near a step to be different from diffusion on a flat surface and was first studied experimentally by Ehrlich and Hudda[1] and later investigated theoretically by Schwoebel,[2] and more recently by Villain[3] and others.[4–10]

The Ehrlich-Schwoebel (ES) barrier E_B is defined as the difference between the activation energy E_{ST} for diffusion of an atom down a step and the energy E_a for diffusion on a flat surface. The presence of a finite positive step barrier has been recently used to explain a variety of experimental results, including reentrant layer-by-layer growth with decreasing temperature in Pt/Pt(111) deposition,[4,5] as well as the observation of large elongated mounds in GaAs deposition on GaAs (100).[10,11] The ES barrier has also been suggested[9] as a possible explanation for the existence of a pyramidlike surface profile and low-index faceting in Cu/Cu(100) deposition.[12] However, despite the recognized importance of the ES barrier in understanding and controlling MBE growth, the magnitude (and sign) of the barrier is not known in general and has been theoretically calculated for only a limited number of systems.[13,14]

In this paper, we estimate the Ehrlich-Schwoebel barrier for Fe/Fe(100) deposition by comparing the results of realistic kinetic Monte Carlo simulations with experimental results[15,16] for the surface width, layer densities, scanning tunneling microscopy (STM) images of surface morphology and out-of-phase reflection high-energy electron diffraction (RHEED) oscillations in the first few monolayers (ML) of growth. We obtain excellent agreement with the experiments by including a finite positive Ehrlich-Schwoebel barrier in our model. In addition, our results appear to resolve a recent controversy[16–19] regarding the existence of a finite step barrier for this system. Surprisingly, we find that from our best estimates, the ES barrier for Fe/Fe(100) is quite small ($E_B = 0.03$–0.06 eV) in comparison with the activation energy for normal diffusion (0.454 eV). This is in marked contrast to previous estimates for fcc (111) metal surfaces[13,14] in which values of the barrier of the order of, or much higher than the normal activation energy were estimated. This indicates that using a simple but realistic model, which captures the essential features of a system in conjunction with experimental data, it is possible to obtain accurate estimates of the interlayer diffusion barrier.

Our study of Fe/Fe(100) deposition has been partly motivated by the availability of detailed experimental results,[15,16] which have already made it possible to quantitatively compare with simulations[20–22] in the submonolayer regime. In particular, using a realistic model with few parameters, we have obtained excellent agreement for the scaling and evolution of the submonolayer island-size distribution in Fe/Fe(100) deposition at temperatures from $T = 20\,^{\circ}\mathrm{C}$ to $356\,^{\circ}\mathrm{C}$.[22] Here we employ a similar model on a square lattice corresponding to the square lattice of adsorption sites on the Fe(100) surface in order to determine the ES barrier in Fe/Fe(100) deposition. We note that our estimate should be regarded as an effective barrier since one expects the barrier to depend on the local configuration. In particular, one expects different values along a straight edge and at a kink. Thus, our estimate for E_B may be considered to be a weighted average over the possible modes of interlayer diffusion. We note, however, that for Fe/Fe(100) the square island morphology implies that a large fraction of step sites will correspond to a straight edge.

In our model, monomers deposited on the flat substrate are assumed to diffuse (nearest-neighbor hops) with diffusion rate $D = D_0 e^{-E_a/k_B T}$; the activation energy for monomers, which attempt to go down a step, is given by $E_{ST} = E_a + E_B$ (with the same prefactor D_0). In addition, the activation energy for atoms with one nearest-neighbor surface atom (bond) is given by $E_1 = E_a + \Delta E_1$. For Fe/Fe(100), the monomer diffusion activation energy $E_a = 0.454$ eV was previously determined experimentally[15] from the scaling of the island density as a function of temperature; an approximate

value of the prefactor D_0 ($D_0 \simeq 7.2 \times 10^{-4}$ cm^2/sec) was determined by comparing the island density with rate equations. By directly comparing our simulation results for the island density at $\theta = 0.07$ ML coverage (for which the effect of the interlayer diffusion barrier is negligible) and $T = 356\,^\circ$C with experimental results, we have also previously estimated $\Delta E_1 \simeq 0.6$ eV,[22] in good agreement with a previous estimate.[16] This large value of ΔE_1 implies that over the temperature range of our simulations (20–250 °C), the probability of diffusion for an atom with more than one in-plane bond is negligible and therefore for simplicity we assume that atoms with two or more nearest-neighbor bonds cannot diffuse. This also implies that the probability of one-bond detachment is negligible at room temperature, in agreement with the observation of a critical island size of $i = 1$ at this temperature.[15] Consequently, at room temperature, the only relevant parameters (other than the deposition rate which is known) in our model are the step barrier E_B and the monomer diffusion rate D. By comparing the island density at $\theta = 0.07$ in our simulations with that obtained experimentally, we have obtained corrected values for D and D_0, which are somewhat less than estimated in Ref. 15.

Since in the experiments the islands are essentially square at room temperature,[15] we have also included an additional parameter $E_e = E_a + \Delta E_e$ corresponding to the activation energy for enhanced diffusion of atoms with one nearest-neighbor along the edge of an island. The existence of such enhanced edge diffusion is consistent with the experimental evidence[15] indicating negligible adatom detachment at room temperature in Fe/Fe(100) along with the observation of square islands in fcc (100) metal epitaxy.[15–17] While the edge-diffusion parameter does not directly affect the island density and size, it strongly affects the island morphology and step density and the probability that an atom on top of an island will encounter a step.[5] Therefore, we have chosen values of the edge-diffusion energy, which provide close agreement with the square island morphology seen at room temperature. Interestingly we find that a very low value of the effective edge-diffusion energy in our model ($\Delta E_e \simeq 0.1$ eV) is necessary to reproduce the observed square morphology at $\theta = 0.07$ and at $\theta = 5.254$ at room temperature.

In order to determine the step barrier E_B for Fe/Fe(100), we have used our model to calculate the root-mean-square surface width $w = [\langle (h - \langle h \rangle)^2 \rangle]^{1/2}$, where h is the surface height in ML, the RHEED intensity $I_R(\theta)$ in the kinematic approximation ($I_R = |\frac{1}{N}\sum_l e^{i\phi h_l}|^2$, where ϕ is the phase difference between layers, N is the number of sites, and h_l is the height in layers of the lth site), and the layer distribution $L(n)$ [where $L(n) = \theta(n) - \theta(n+1)$ and $\theta(n)$ is the coverage in the nth layer] as a function of temperature and the coverage θ for a range of values of E_B. We have compared our results with the experimental data.[15,16] We have also compared the surface morphology from simulations with that found in the experiment.

Figure 1 shows a comparison between our results for the surface width ($\Delta E_e = 0.1$ eV) and experimental results at room temperature,[15] determined from STM cross

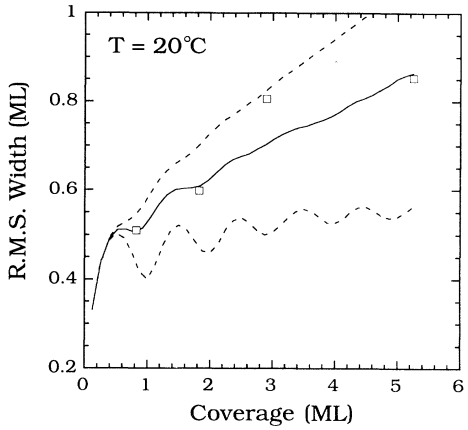

FIG. 1. Root-mean-square surface width for Fe/Fe(001) deposition from simulations with $\Delta E_e = 0.1$ eV, $E_B = 0.03$ eV (solid line), $E_B = 0.0$ eV (lower dashed line), and $E_B = 0.04$ eV (upper dashed line) at room temperature along with experimental results (open squares) at a deposition rate of 0.0257 ML/sec.

sections after deposition. As can be clearly seen, without a step barrier ($E_B = 0.0$) there are pronounced oscillations while the surface width is significantly smaller than found in the experiment. On the other hand, with a finite ES barrier ($E_B \simeq 0.03$ eV) we find reasonable agreement with the experimental results. Furthermore, a slight increase in the step barrier ($E_B = 0.04$ eV) leads to values for the surface width that are larger than in the experiment. This indicates that the step barrier while quite small is finite, while the room temperature surface width appears to be a very sensitive indicator of the step barrier magnitude. We note that by varying the edge-diffusion energy as well as taking kinetic restructuring effects into account,[23] we have obtained upper and lower bounds for E_B. For example, lowering the edge-diffusion barrier to 0.05 eV (leading to islands that are more regular than observed in experiment) gives $E_B \simeq 0.02$ eV, indicating a lower bound for E_B of approximately 0.03 eV. On the other hand, including local restructuring of freshly deposited atoms to nearest-neighbor sites (transient kinetics) (Ref. 23) serves to reduce the effectiveness of the barrier and gives an upper bound of $E_B \simeq 0.06$ eV. Thus, by studying the limiting cases of our model, we have obtained strong bounds on E_B using the width data.

In order to confirm our estimate of the step barrier based on the width data, we have also compared our results for the RHEED intensity with those obtained in experiments at room temperature and above. Figure 2(a) shows the experimental results for the out-of-phase RHEED intensity obtained in Ref. 15 (10-keV beam, $\Theta = 64 \pm 4$ mrad) at $T = 20$–$250\,^\circ$C. The corresponding simulation results for the kinematic RHEED intensity I_R with $E_B = 0.03$ eV for the same temperatures and deposition rate[24] are shown in Fig. 2(b). As can be seen, the behavior of the RHEED oscillations is qualitatively quite similar to the experiments. In particular at $T = 20\,^\circ$C, the RHEED oscillations are significantly damped in agreement with the experimentally observed behavior. In contrast, the corresponding simu-

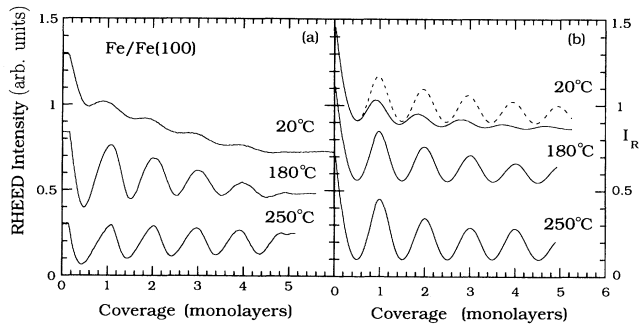

FIG. 2. (a) Experimentally observed out-of-phase ($\phi \simeq 3\pi$) RHEED intensity (Ref. 15) for $T = 20, 180$, and 250 °C. (b) Simulation results for the kinematic RHEED intensity I_R with ($E_B = 0.03$ eV, solid lines) and without ($E_B = 0.0$ eV, dashed lines) a step barrier.

lation results for $E_B = 0.0$ eV disagree with the experimental data and show much weaker damping. Similarly, at $T = 180$ °C, the RHEED oscillations for the simulated data with $E_B = 0.03$ eV are only slightly damped in agreement with the experiment; at $T = 250$ °C, the oscillations are essentially undamped beyond the second oscillation, again in agreement with the experiments. Therefore, our estimates of the step barrier using the RHEED intensity data are quite consistent with the results obtained from the width data. We should point out, however, that the exact calculation of the out-of-phase RHEED intensity from experimental morphology is quite complicated due to such effects as multiple scattering and scattering from step edges. We also note that in the past, crude models have yielded good agreement with RHEED data. Therefore, a more quantitative comparison between the STM results for the surface structure and our simulation results (described below) provides a more stringent test of our model. We note that at $T = 180$ and 250 °C the kinematic RHEED results for $E_B = 0.0$ eV (not shown) and $E_B = 0.03$ eV are essentially the same since at these high temperatures, the effects of the barrier are much less noticeable.

In order to further test our estimate of the effective barrier E_B as well as the validity of our model, we have also compared the surface morphology obtained from our simulations (Fig. 3) with the experimental STM results.[15] As can be seen from the gray-scale plot of our simulation results in Fig. 3, at $T = 20$ °C ($\theta = 5.254$ ML) the fifth layer is only partially complete, and contains a large number of small slightly irregular square islands with a significant number of additional islands in the sixth layer. On the other hand, at $T = 180$ °C ($\theta = 4.93$ ML), the islands in the fifth layer are much larger and almost percolate across the entire system, while there is still a significant amount of growth in the sixth layer. Finally, at $T = 250$ °C ($\theta = 4.9$ ML), the fifth layer is almost complete while the fraction of islands in the sixth layer is much smaller. These features are all in good qualitative agreement with the STM micrographs shown in Ref. 15 and indicate that the presence of a small, but finite step barrier is needed to capture the essential features of the surface morphology in Fe/Fe(100) deposition.

This qualitative agreement is further supported by the quantitative comparison between our simulation results for the layer distribution and the experiments shown in Fig. 4 for $T = 20$ °C and $T = 250$ °C. As can be seen, at room temperature and at $T = 250$ °C, there is good agreement between our simulation results for the layer distribution with a step barrier included and the experimental results for the layer distribution. In contrast, for the case of zero step barrier ($E_B = 0.0$), there is relatively poor agreement with the experimental results at $T = 20$ °C. Thus, we find good agreement with all the experimental data by assuming a small but finite value of the ES barrier between 0.03 and 0.06 eV.

In summary, we have developed a realistic yet simple model of the early stages of growth in Fe/Fe(100) deposition. By including a finite value of the interlayer diffusion barrier, we have obtained good agreement with the experimental results for the RHEED oscillations, surface width, surface morphology, and layer distributions at $T = 20$, 180, and 250 °C.

Our calculation of the step barrier for iron may also help to resolve an existing controversy regarding multilayer growth in Fe/Fe(100). In particular, a recent

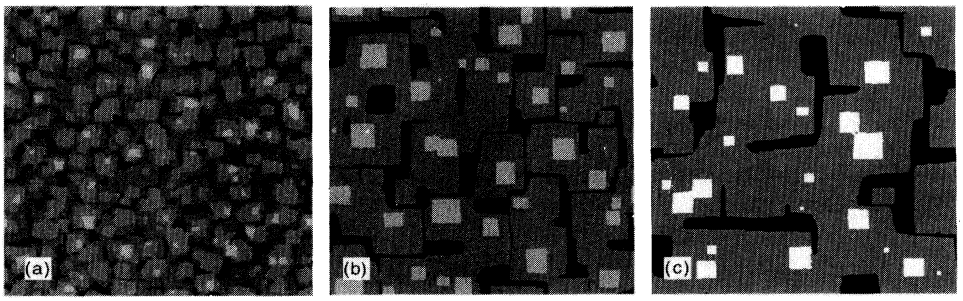

FIG. 3. Gray-scale pictures of the surface morphology obtained from simulations with $E_B = 0.03$ eV at (a) $T = 20$ °C, $\theta = 5.254$ ML, and $L = 210$; (b) $T = 180$ °C, $\theta = 4.93$ ML, and $L = 418$; and (c) $T = 250$ °C, $\theta = 4.9$ ML, and $L = 418$, where L is the system size in units of the lattice spacing and deposition conditions are the same as in Fig. 2 of Ref. 15. Lighter shades of gray correspond to increasing surface height. The system size in (a) is the same as Fig. 2(a) of Ref. 15 while in (b) and (c) the system size is slightly larger than half that of corresponding pictures in 15.

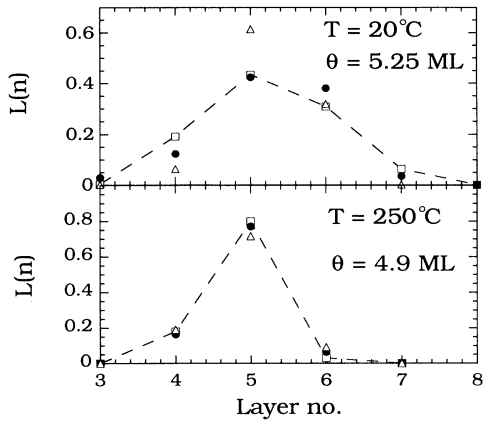

FIG. 4. Experimental (open squares connected by dashed lines) and simulated ($E_B = 0.03$ eV, filled circles; $E_B = 0.0$ eV, triangles) layer distribution $L(n)$ at $T = 20\,^{\circ}$C and $250\,^{\circ}$C.

diffraction study by He *et al.*[18] found self-affine scaling behavior, which may be interpreted as corresponding to a negligible ES barrier. However, recent STM and structure factor measurements by Stroscio *et al.*[15,19] indicate a surface morphology consisting of mounds with a typical length scale. Our estimates of a finite but small positive barrier lend strong support to the conjecture[19] that a barrier is responsible for the mounds in this system.

Surprisingly, our estimate for the step barrier is much smaller than the activation energy for diffusion on a flat surface. As already noted, this is in marked contrast with previous estimates for fcc (111) metal surfaces[13,14] in which values of the barrier of the order of, or much higher than the normal activation energy were estimated. While future studies using a larger number of interaction energies and a more detailed geometry and kinetics may be able to refine our estimate of the ES barrier and determine the mechanism for interlayer diffusion, we believe that our model has captured the essential features of Fe/Fe(100) in the first few layers of growth.

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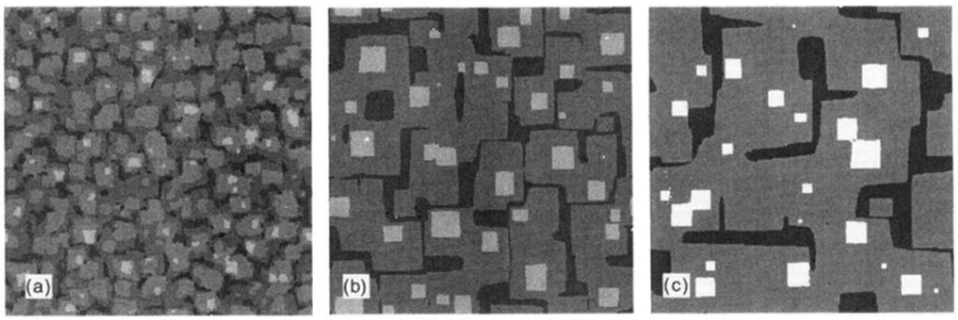
[24] The kinematic RHEED intensity has been calculated slightly off the exact out-of-phase condition, in accordance with the error bars (±4 mrad out of 64 mrad) on the RHEED measurement.



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