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Impact of STM tip on coarsening of In atomic chains on Si(100): A kinetic Monte Carlo study

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ABSTRACT

Using a Kinetic Monte Carlo approach, we investigate the potential influence of STM tip on nucleation and subsequent growth of single-atom wide In chains on Si(100) during room-temperature deposition. Specifically, we examine the effect of repeated scanning on the stability of odd-size chains, as well as coarsening of chains in the *post-deposition* regime. In our simulations, the effects of the strong field developed between the STM tip and the positively charged In adsorbates when the sample is positively (negatively) biased are modeled by slightly lowering (raising) the activation barriers of adatoms directly underneath the tip. Evidence of stochastic ripening – a coarsening mechanism relying on a series of adatom exchanges between adjacent islands in one-dimensional systems and not driven by a difference in the chemical potential – will be presented. Finally, we will show that coarsening may be disrupted by STM tip effects.

1. Introduction

Owing to its versatility, the scanning tunneling microscope (STM) has found widespread use in either imaging surfaces or modifying them since its introduction by Binnig et al. [1] in the mid-eighties. In particular, the latter function has been employed to actively manipulate adsorbates with great precision. When STM is used to image surfaces, however, it is generally assumed that the effect on the surface is minimal under a certain range of operating conditions. Hence, studies that quantify the influence of STM tips are rare. To our knowledge, there is no study that correlates impact of STM tip with tunneling conditions. Further complicating the picture is that unless the influence of STM tip is significant enough to cause easily identifiable modifications on the surface, STM images are obtained with STM tip effects already built-in. The challenge is then to determine what the surface might look like without these effects.

Previous studies [2,3] on the influence of STM tip on migration of In adatoms on Si-based surfaces suggest slight positive charging of In atoms upon adsorption due to a charge transfer between adsorbate and the surface, accompanied by the creation of static dipoles. This partial polarization of the adsorbate thus suggests a differentiation on the impact of the STM tip on positively biased versus negatively biased samples. For example, a study on In adatom migration on Si(111)-$\sqrt{3} \times \sqrt{3}$ reported a localized increase (decrease) in In coverage under the tip when the bias voltage is negative (positive) [2]. For In/Si(100), Kocan et al. [3] reported that when negatively biased, In adatoms directly underneath the STM tip exhibit greater "liveliness", i.e., adatoms detach and attach more readily than if the sample were positively biased. Previously, there had been indications that field-driven migration as a result of adsorbates' polarization and their subsequent interaction with the tip can also be found in related systems. One such system is Tl/Si(100) in which polarized Tl adatoms migrate under the influence of STM tip [4].

The above qualitative observations imply that if a large enough area of the sample can be monitored, it should show a mass transfer between the region close to the tip and that just outside of it in which the net flow direction is dictated by the tip bias. In this work, we ask if under repeated scans, such a net migration of adatoms could be replicated on a bigger scale involving scanned and unscanned regions. Additionally, we are interested to know whether such an adatom mass transfer on a "global" scale might leave an imprint on the coarsening process.

In this simulation study, we also aim to take preliminary steps towards quantifying the presence of STM tip effects on the surface morphology. It should be noted that establishing empirical trends relating STM operating conditions, such as tunneling current, bias voltage, as well as scan rate, to key quantities, such as time constants and detachment rates at island edges is hampered by dearth of statistical data from STM measurements, as is readily apparent from reading Ref. [3].

Thus to address these issues, we have developed a kinetic Monte Carlo (KMC) model that simulates In migration on Si(100) and which explicitly incorporates the influence of the STM tip. At the outset, it should be stressed that there is no attempt to include detailed

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information on the STM tunneling conditions, like the magnitudes of the tunneling current and the bias voltage, into the model. Rather, noting that a positively (negatively) biased tip tends to drive (attract) adatoms from the sites directly underneath it, our KMC model can be made to operate in the *repulsive* (*attractive*) mode through appropriate modifications to the activation barriers between sites centered on the tip. Thus, in this paper the two modes of invasive STM operation, *repulsive mode* and *attractive mode*, correspond to experimental settings in which the tip is negatively and positively biased, respectively under the assumption that In adatom acquires slight positive charging upon adsorption.

This paper is organized as follows: In Section 2, we provide details of the KMC model that mimics formation In atomic chains that is typically present on Si(100) during room-temperature deposition. This model necessarily includes implementation of STM tip effects via modification of the diffusion barriers on sites close to the tip. In Section 3, we examine the impact of scanning on the stability of odd-sized chains by looking at the detachment rates at island edges. Additionally, we calculate the coarsening exponents, with or without scanning and compare them with those associated with conventional ripening mechanism. In Section 4, we conclude the article by summarizing our main findings.

2. Methodology

2.1. Growth model details

Incorporating features established through various experimental and theoretical studies, the self-assembly of one-dimensional In islands, or chains, on flat Si(100) surface during room-temperature deposition was simulated using KMC techniques in a prior work [5]. One of the most noteworthy of these features as revealed from STM images is that In adatoms tend to nucleate around *C-defect* sites [3]. Growth then proceeds through so-called *surface polymerization reaction* along the direction orthogonal to the Si dimer row, a characteristic common among other group III metals on Si(100) [6,7]. Furthermore, growth is considered reversible as atoms were observed to continuously attach and detach from the "free" end of the chain that is not bound to a defect [3].

In this work, we will incorporate the effect of the STM tip by treating it as a highly mobile, localized distortion in the potential energy landscape that either repels or attracts adatoms and small clusters in its vicinity, depending on the bias voltage between the tip and the sample. Specifically, we will model the presence of the tip by modifying the diffusion barriers for adatom hops along the scan direction involving sites directly under the tip and its two nearest neighbors on either side (sites 1, 2 and 3). See Fig. 1. This feature mimics the effect of the strong electric field developed between the indium adatoms (which upon adsorption acquire a net positive charge) and the tip.

2.2. Model parameters and details of implementation

The KMC model used in the present work is an expanded (and modified) version of the original atomistic lattice-gas model introduced in Ref. [5], in which the Si(100)-2 × 1 surface is represented by a rectangular grid consisting of $N_1 \times N_2$ sites. Each point on the grid coincides with a Si dimer and corresponds to a minimum on the potential energy surface (PES) map; each of these sites are stable binding sites for In adatoms. For In/Si(100), the orientation of the In dimer, the basic building block of the atomic chains, is governed by the *parallel-dimer* model [6,7]. According to this model, the atomic chains runs perpendicular to the Si dimer row; thus, defining the direction of the metal row. (Si dimer rows and metal rows are mutually perpendicular). Below, we summarize the key aspects of our model.

(a) Random distribution of *C-defects*. Prior to In deposition, very low concentration of *C-defects* ~10^{-3} (per site) are distributed uniformly on the bare Si(100) surface. Two adjacent defects are explicitly disallowed.

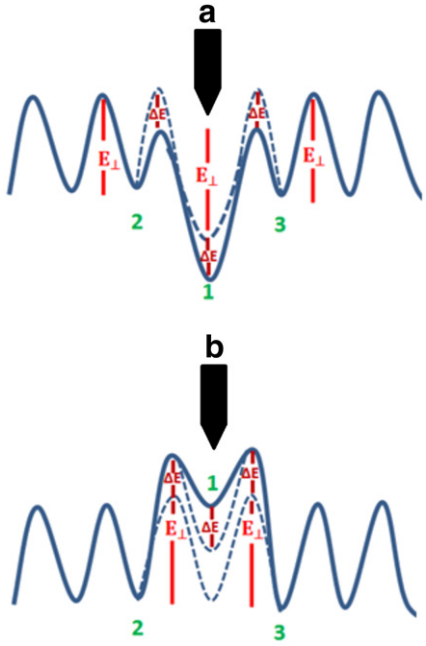

Fig. 1. (a) *Attractive mode*. In this mode, the activation barrier for hopping to site 1 (the site underneath the tip) from sites 2 and 3 is lowered by ΔE, while the reverse hop is raised by the same amount. (b) Repulsive mode. In this mode, the activation barrier for hopping to site 1 (the site underneath the tip) from sites 2 and 3 is raised by ΔE, while the reverse hop is lowered by the same amount.

(b) Random In adsorption. In atoms are randomly deposited on available sites, provided the site is empty and not laterally adjacent to an island or an adatom. In addition, atoms may not deposit on *C-defect* sites. The adsorption rate is given by $F = 1 \times 10^{-4}$ ML/s, typical values used in the experiments [3,8].

(c) Adatom diffusion. Adatoms hop along the two principal directions (parallel and perpendicular to the Si dimer row). Their hop rates, denoted by h, are presumed to obey Arrhenius law, $h = \nu\, e^{E/kT}$. Here, ν is the hopping frequency (also known as the prefactor) whose value ranges from 10^{12} to 10^{13} s^{-1} (though a recent estimate indicates that it could be as low as 4×10^{11} s^{-1} [9]), E is the activation barrier, while T is the temperature. In the case of In/Si(100), diffusion is almost isotropic [5,9], thus justifying the use a common diffusion barrier $E = 0.27$ eV along the two principal directions in our simulations. Furthermore, the same restriction on the possible adsorption sites imposed in (b) while trying to deposit adatoms are also applied here.

(d) Island nucleation and growth. In homogeneously nucleated islands, two free adatoms that hop on adjacent sites along the same metal row (perpendicular to the Si dimer row) form a chain or island. On the other hand, an adatom landing next to a *C-defect* is trapped, forming a heterogeneous island in the process. Subsequent growth proceeds along the direction perpendicular to the dimer row.

(e) Reversible detachment and island decay. End atoms of an island detach at rates indicated in Table 1. For very small chains, such a rapid detachment may ultimately lead to dissolution.

(f) Bortz–Kalos–Lebowitz (BKL) algorithm. Our KMC simulations employ the highly efficient BKL algorithm. Initially, an exhaustive list of allowed processes (as well as a secondary list of associated adatoms) is produced. Then, a random number is drawn in order to choose which process to implement; processes with the greatest assigned weight are chosen more often than the other processes. Once a process is selected, another random number is selected in order to choose the particular adatom that will execute the process. The final step involves updating the list of processes (and adatoms). According to this method,