

# Monte Carlo derived diffusion parameters for Ga on the GaAs(001)-(2×4) surface: A molecular beam epitaxy–scanning tunneling microscopy study

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The migration of individual Ga atoms on the technologically important GaAs(001)-(2×4) reconstructed surface has been studied as a function of substrate temperature and As<sub>4</sub> pressure using a combined molecular beam epitaxy and scanning tunneling microscope ultrahigh vacuum multichamber facility. We have deposited 10% of a plane of Ga onto a GaAs(001) surface with a low defect density (<1% ) and with large terraces (>0.5 μm) to avoid the influence of surface defects like step edges and vacancies. Both the island number density and the geometry are measured and compared to Monte Carlo solid-on-solid simulations. Basic diffusion parameters, such as the activation energy, directional hopping-rate ratio, directional sticking-probability ratio, etc., are reported. © 2000 American Vacuum Society. [S0734-2101(00)08204-X]

## I. INTRODUCTION

The optical properties of III–V compound semiconductors are making high-speed global, wireless communications possible.<sup>1</sup> This application has created a demand for higher performance device structures, which are more complex and more difficult to realize. Unlike silicon-based devices, which are primarily fabricated using ion implantation,<sup>2</sup> III–V structures are formed at the surface by depositing one plane of atoms on top of another until the entire structure is formed. A more accurate atomic-scale understanding of the fundamental physics governing the motion of group III and group V atoms on III–V crystal surfaces would aid the fabrication process.

To date, several macroscopic methods, primarily reflection high-energy electron diffraction (RHEED), have been used to study Ga diffusion on the GaAs(001) surface.<sup>3–9</sup> In some of these studies, an estimate of the surface adatom diffusion length at one temperature is obtained from knowing the average terrace width at which the growth mode changes from two-dimensional island nucleation to step-flow mode. Interpretation of these experiments can be ambiguous because the influence of surface defects such as vacancies, step bunching, and interactions between adatoms themselves cannot be taken into account. Furthermore, since the electron beam averages over the entire substrate, temperature gradients and morphological variations across the wafer will effect this type of measurement. Some of these RHEED studies have utilized scanning tunneling microscopy (STM) to image the morphology as a function of multiple monolayers of coverage.<sup>5,10</sup> However, growing more than about 10% of a monolayer on a surface that is not flat, makes it difficult to separate the basic diffusion process from the large number of other interactions that can occur.

First-principles total-energy calculations of the pure migration of Ga atoms on the GaAs(001)-(2×4) reconstructed

surface have been performed.<sup>11</sup> These calculations predict an activation barrier of 1.2 eV along the As dimer rows (i.e., along the [1 $\bar{1}$ 0] direction) and 1.5 eV across them (i.e., along the [110] direction). These state-of-the-art calculations are only true for the pure migration of individual Ga atoms on an otherwise perfect (2×4)-reconstructed surface in the absence of defects and an arsenic flux. These predictions have not been tested experimentally to date. The recent application of STM to the study of atomic diffusion on various elemental single crystal surfaces (e.g., Si and Fe) has significantly broadened the fundamental knowledge of the motion of atoms on these surfaces.<sup>12–15</sup> STM studies of diffusion on the GaAs(001) surface have also been carried out under metalorganic vapor-phase epitaxy conditions.<sup>16,17</sup> Unfortunately, these GaAs(001) diffusion studies do not provide an accurate test for the above theoretical predictions, since many other processes, like organic chemical reactions, are involved in the growth. Nucleation and growth on the molecular beam epitaxy (MBE)-prepared GaAs(001) surface has been experimentally investigated using STM and kinetic Monte Carlo simulations.<sup>18</sup> This study deposits a submonolayer amount of material and fits the island's geometry to simulations. Unfortunately, the study is only done at one temperature, making it difficult to address the diffusion properties.

Motivated by these issues, the activation energy for diffusion of gallium on the GaAs(001)-(2×4) reconstructed surface has been measured under an ultrahigh vacuum molecular beam epitaxy growth condition. Both the GaAs island number density and island geometry are measured from STM images for samples after depositing 10% of a plane of Ga atoms onto the GaAs(001)-(2×4) reconstructed surface held at various temperatures and exposed to different As<sub>4</sub> fluxes. This measurement is performed with atomic-scale resolution, naturally allowing for the influence of steps, vacancies, and other defects to be accounted for, further isolating the pure activation energy of diffusion. One could apply rate equation analysis to this data to determine the diffusion coefficient and activation barrier for isotropic two dimen-

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sional motion or one dimensional motion.<sup>19,20</sup> However, by applying modeling using Monte Carlo simulations one can obtain a more detailed picture of the atomic process.<sup>12</sup> In this study, we apply Monte Carlo analysis and obtain information about the activation barrier for Ga to hop from one site to the next, the relative hopping rates in perpendicular directions on the surface, the relative sticking rates to existing islands when approached from perpendicular directions, and finally we gain insight into the role that arsenic plays in altering the Ga diffusion and growth process.

## II. EXPERIMENT

Experiments were carried out in an ultrahigh vacuum (UHV) multichamber facility ( $5-8 \times 10^{-11}$  Torr throughout) which contains a solid-source MBE chamber (Riber 32P) that includes a substrate temperature determination system accurate to  $\pm 2$  °C.<sup>21</sup> This chamber is connected to a surface analysis chamber with an STM (Omicron).<sup>22</sup> Commercially available, “epiready,”  $n+$  (Si doped  $10^{18}/\text{cm}^3$ ) GaAs(001)  $\pm 0.1^\circ$  substrates were loaded into the MBE system without any chemical cleaning. The surface oxide layer was removed and a 1.5- $\mu\text{m}$ -thick GaAs buffer layer was grown at 580 °C using a growth rate of 1.0  $\mu\text{m}/\text{h}$  as determined by RHEED oscillations and an As<sub>4</sub> to Ga beam equivalent pressure (BEP) ratio of 15.

An algorithm was developed that produces a well ordered GaAs(001)-(2 $\times$ 4) reconstructed surface with 0.5–1  $\mu\text{m}$  wide terraces essentially free of defects.<sup>19</sup> This algorithm consists of first annealing the substrate at 600 °C with an As<sub>4</sub> BEP of 1.0  $\mu\text{Torr}$  for 15 min. This procedure eliminates step bunching, yet leaves each terrace with 1 monolayer (ML) high adatom and vacancy islands. Another 15 min anneal at 570 °C with the same As<sub>4</sub> BEP eliminates these islands and leaves the surface flat.

Ga diffusion occurs on this flat, well-ordered GaAs(001)-(2 $\times$ 4) reconstructed surface consisting of two adjacent parallel As dimers with a third adjacent parallel As dimer 1 ML below the other two.<sup>23</sup> Numerous samples were prepared by depositing 10% of a plane of Ga at a specified temperature, while simultaneously exposing the surface to two different As<sub>4</sub> BEPs. In order to lock in the as-grown structure, we first start quenching the sample from 570 °C at 1.5 °C/s. While the sample was cooling, enough As<sub>4</sub> flux was maintained on the sample to avoid degradation of the surface. Once the sample temperature reached the desired value, the Ga shutter was opened for 0.5 s to deposit 10% of a plane of Ga while the As<sub>4</sub> BEP was simultaneously increased to either 1.0 or 10.0  $\mu\text{Torr}$ , (i.e., the growth rate is set to 20% of a plane per second). A timing chart for this deposition has been published elsewhere.<sup>20</sup> Finally, the sample temperature was held at 450 °C for 15 min to allow any As<sub>4</sub> inside the growth chamber to be pumped out and not condense on the surface of the sample. It has been determined that this anneal does not affect the island geometry or number density.

As mentioned earlier, samples could not be prepared for any temperatures above 570 °C because the surface would

spontaneously roughen when annealed above this temperature.<sup>24</sup> In addition, the experiments could not be performed for substrate temperatures below 460 °C because the surface reconstruction will transform from the (2 $\times$ 4) to the  $c(4 \times 4)$ . The As<sub>4</sub> BEP of 1.0  $\mu\text{Torr}$  and the Ga deposition rate of 20% of a plane per second results in an As<sub>4</sub>/Ga flux ratio of  $\sim 15$ , thereby recreating typical GaAs growth conditions which use a ratio between 10 and 20. When the As<sub>4</sub> BEP is 10.0  $\mu\text{Torr}$ , the As<sub>4</sub>/Ga flux ratio is  $\sim 150$ , allowing the study of Ga diffusion under extremely high arsenic concentration conditions.

After the deposition and the 15 min anneal at 450 °C, the sample was cooled to room temperature, transferred to the STM without breaking UHV, and imaged at room temperature. For each sample, 6–12 200 nm  $\times$  200 nm or 100 nm  $\times$  100 nm filled-state STM images were acquired using tips made from single crystal  $\langle 111 \rangle$ -oriented tungsten wire, a sample bias of  $-3.0$  V and a tunneling current set point of 0.05–0.2 nA. For each sample, an average island number density was determined from 6–12 STM images.

To quantify the diffusion process, Monte Carlo simulations of the MBE growth process were completed. These simulations use a solid-on-solid model, where single adatoms are randomly dropped onto an area of 200 $\times$ 200 lattice sites until a total coverage of 0.1 ML is reached. These deposited adatoms model the Ga adatoms deposited during the MBE growth. The simulation assumes that a single free adatom will diffuse indefinitely until either two adatoms collide and stick (nucleating an island) or the adatom collides and sticks to an existing island (growing an island). Once the adatom is stuck, it is excluded from further diffusion. Therefore, the critical nucleation size is two adatoms and the probability of three or more adatoms simultaneously colliding is considered negligible. The adatoms are assigned a jump rate  $J$ , which is related to the diffusion rate  $D$  via the lattice constant,  $a$  (i.e.,  $J=4D/a^2$ ). Simulations were performed which varied the jumping rate in decade-size steps from 1 to  $10^9$  Hz. For each jump rate, the probability of hopping in perpendicular directions (directional hopping-rate ratio) was varied from (1 to 1) to highly anisotropic (1 to 1000). For each of these simulations, the probability of sticking to other atoms when approached from different directions (directional sticking-probability ratio) was varied from equal (1 to 1) to highly anisotropic (1 to 1000 and 1000 to 1).

## III. RESULTS

### A. 1.0 $\mu\text{Torr}$ As<sub>4</sub> BEP data and Monte Carlo results

Characteristic STM images of the GaAs surface after the deposition of 10% of a monolayer at three successively higher temperatures are shown in Figs. 1(a), 1(b), and 1(c). Each of these images measures 100 nm $\times$ 100 nm and was cropped far from the edges of a much larger terrace. The lower temperature result shown in Fig. 1(a) has a larger number of smaller size islands when compared to the other two temperatures shown in Figs. 1(b) and 1(c). This is observed because the island growth rate decreases while the nucleation

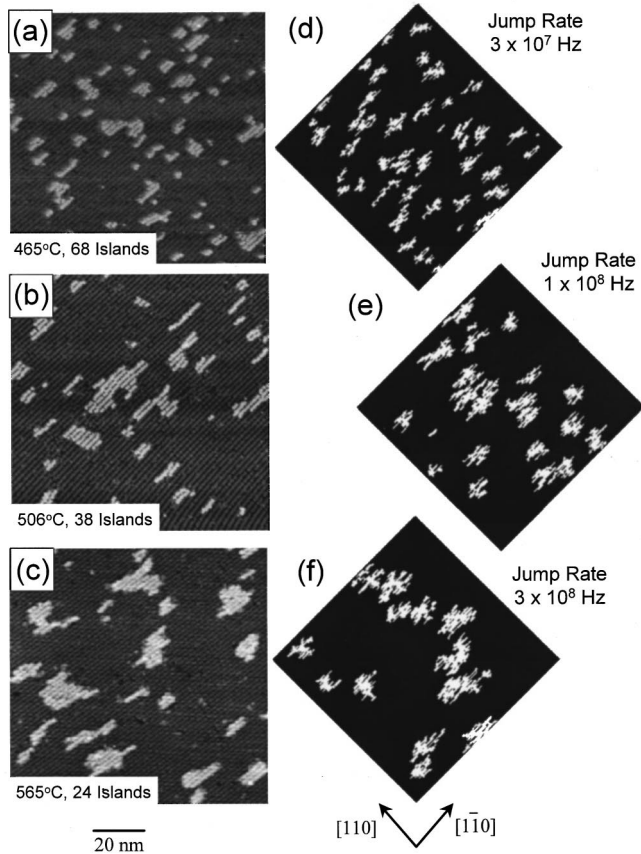


FIG. 1. (a)–(c) Filled state STM images taken at a sample bias of  $-3.0$  V and a demanded tunneling current of  $0.2$  nA of the GaAs(001)-(2 $\times$ 4) reconstructed surface after deposition of 10% of a monolayer of Ga under a  $1.0$   $\mu$ Torr  $\text{As}_4$  flux at the temperatures indicated. The islands are 1 ML high (0.28 nm). For comparison, (d)–(f) are simulated STM images from random walk computer simulations as a function of the jump rate using a directional hopping-rate ratio of 100:1 ( $[\bar{1}\bar{1}0]:[110]$ ) and directional sticking-probability ratio of 100:1 ( $[\bar{1}\bar{1}0]:[110]$ ).

rate of new islands increases when the temperature is lowered. With more islands on the surface they must be smaller in size because the coverage for each sample is fixed. The islands formed under these conditions are not circular, but favor elongation along the  $[\bar{1}\bar{1}0]$  direction.

To compare with the STM images, hundreds of Monte Carlo simulated STM images were computed. Both the number density of the islands and the geometry of the islands (i.e., aspect ratio) in the STM images were fit using three parameters in the Monte Carlo simulations: jumping rate, directional hopping-rate ratio (hop-ratio), and directional sticking-probability ratio (stick ratio). A higher jump rate favors island growth over island nucleation. Therefore, increasing this parameter predominately results in a smaller number of islands. Increasing the anisotropy of the hop ratio predominately lowers the island number density. Increasing the anisotropy of the stick ratio predominately alters the geometry of the islands. If the geometry of the islands is not considered, the stick ratio can be adjusted such that a unique set of hop ratio versus jump rate parameters cannot be obtained. However, a stick ratio of 1 to 1 or 1000 to 1 results in

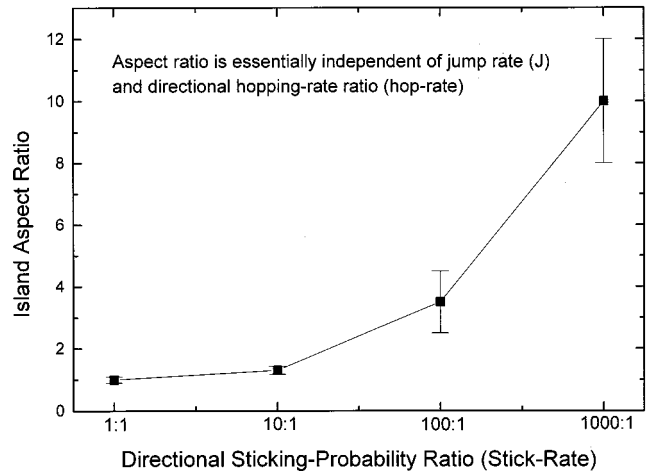


FIG. 2. Plot of the island aspect ratio vs directional sticking-probability ratio from the Monte Carlo simulations. The aspect ratio is essentially independent of the jump rate  $J$  and the directional hopping-rate ratio. The slight dependence on these latter two parameters is accounted for in the error bars.

an island geometry that is not observed, thus making this an important fitting parameter. Our algorithm for finding the best set of fitting parameters began with noticing that the aspect ratio of the islands measured with STM are essentially constant for all temperatures and arsenic fluxes. It was determined that the geometry of the simulated islands depends primarily upon the stick ratio and is essentially independent of the jump rate and hop ratio as depicted in Fig. 2. Furthermore, a stick ratio of 100:1 ( $[\bar{1}\bar{1}0]:[110]$ ) best fits the observed island geometry. To find the hop ratio, the number density of the simulated islands as a function of jump rate was required to match the number density of the STM imaged islands as a function of temperature. It was determined that a hop ratio of 100:1 ( $[\bar{1}\bar{1}0]:[110]$ ) for all jump rates provided the best fit for the lower arsenic pressure data. Three characteristic simulated STM images measuring  $80$  nm $\times$  $80$  nm, having 10% Ga coverage, and a deposition rate of  $0.2$  ML/s are shown in Figs. 1(d), 1(e), and 1(f).

The average island number densities for all data sets are shown in Fig. 3. The error bars represent the standard deviation in the average island number density counted from all the STM images acquired at that temperature. Certain criteria were used to define an island. First, it has to be 1 ML high (0.28 nm). Second, the monolayer high island must have (2 $\times$ 4) reconstruction features, such as correct positioning relative to underlying dimer rows. These criteria eliminated the small white specks existing directly on top of the dimer rows that were observed on some of the samples [see, for example, Fig. 1(a)], which are thought to be adsorbed  $\text{As}_4$  molecules resulting from condensation inside the MBE chamber after the Ga deposition.<sup>25</sup> However, the inclusion of these features shifted the average island count less than the standard deviation. The average island number density generated from ten different random number seeds as a function of jump rate for the best-fit set of parameters is also shown in Fig. 3 (filled circles). Note, that only the jump rates relevant to the experimental data are shown. The error bars represent

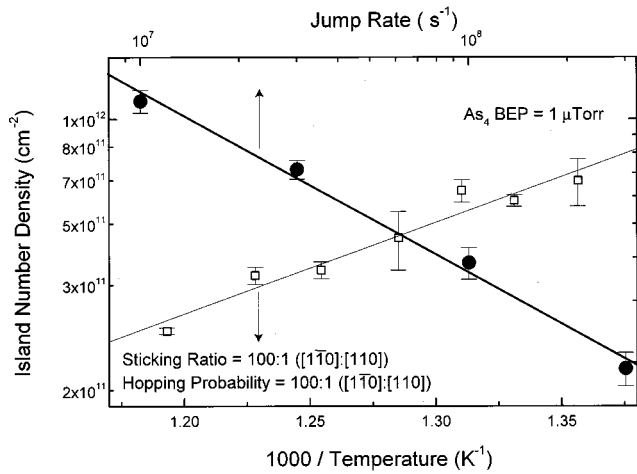


FIG. 3. Measured island number density (open squares) vs the deposition temperature (lower axis) for the  $1.0 \mu\text{Torr}$   $\text{As}_4$  BEP data, with error bars representing  $\pm 1$  standard deviation in the number of islands counted. The island number density (filled circles) vs the jump rate (upper axis) from random walk simulations are also shown. The error bars represent  $\pm 1$  standard deviation in the number of islands counted from ten different random number seeds. The simulations use the directional hopping-rate ratio of 100:1 ( $[1\bar{1}0]:[110]$ ) and directional sticking-probability ratio of 100:1 ( $[1\bar{1}0]:[110]$ ). The line going through the simulation data points is a least-squares linear fit.

the standard deviation in the average island number density counted from the ten simulations.

By combining the Monte Carlo simulation results with the experimental data a diffusion coefficient can be obtained. The jump rate  $J$  for each experimental data point is found by matching the island number density from the measured data to the least squares fit of the simulation. This jump rate is converted to a diffusion rate  $D$  (i.e.,  $D = Ja^2$ ) and plotted versus temperature in Fig. 4. The diffusion rate and temperature are related by  $D = D_0 \exp[-E_a/(k_B T)]$ , where  $k_B$  is Boltzmann's constant,  $E_a$  is the activation energy to hop from one site to the next,  $D_0$  is the prefactor, and  $T$  is the absolute temperature. The activation energy is calculated from a least squares fit to be  $1.0 \pm 0.1$  eV with a prefactor of  $\sim 0.2 \text{ cm}^2/\text{s}$ .

## B. Comparison between the $1.0 \mu\text{Torr}$ $\text{As}_4$ BEP and $10.0 \mu\text{Torr}$ $\text{As}_4$ BEP data

In addition to measuring the island number density as a function of substrate temperature, it was also measured for two different  $\text{As}_4$  pressures during the Ga deposition (i.e.,  $1.0$  and  $10.0 \mu\text{Torr}$   $\text{As}_4$  BEP). Two characteristic STM images acquired at  $465^\circ\text{C}$ , but with a factor of ten difference in the  $\text{As}_4$  gas pressures, are shown in Figs. 5(a) and 5(b). There is about a factor of two decrease in the island number density when the  $\text{As}_4$  pressure is increased by this amount, and this difference persists for all temperatures.<sup>20</sup> In this study, these differences are quantified using the Monte Carlo simulations. Two characteristic simulated STM images measuring  $80 \text{ nm} \times 80 \text{ nm}$ , having 10% Ga coverage, and a deposition rate of  $0.2 \text{ ML/s}$  are shown in Figs. 5(c) and 5(d). The simulation in Fig. 5 (c) was calculated using the

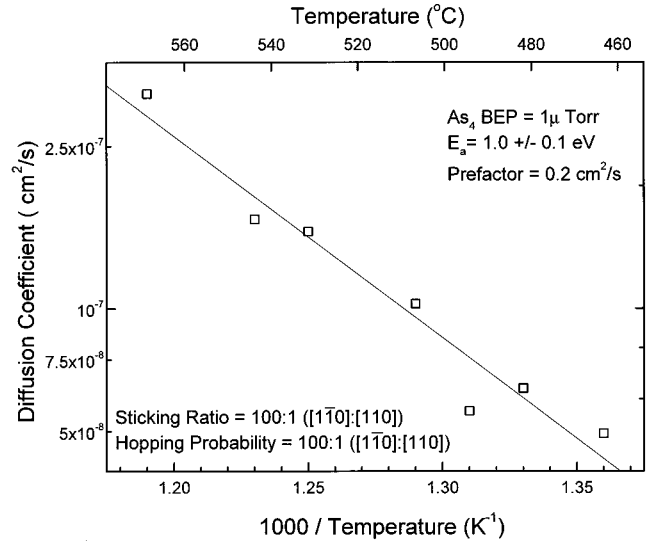


FIG. 4. Diffusion coefficients (open squares) vs temperature obtained by combining the measured experimental island number densities from the  $1.0 \mu\text{Torr}$   $\text{As}_4$  BEP data with the random walk simulations. The least squares fit (solid line) gives an activation energy of  $1.0 \pm 0.1$  eV and a prefactor of  $\sim 0.2 \text{ cm}^2/\text{s}$ .

same conditions as before, while the simulation in Fig. 5(d) used these same parameters except the directional hopping-rate ratio was changed from 100:1 ( $[1\bar{1}0]:[110]$ ) to 5:1 ( $[1\bar{1}0]:[110]$ ). Even though the fit between Figs. 5(b) and

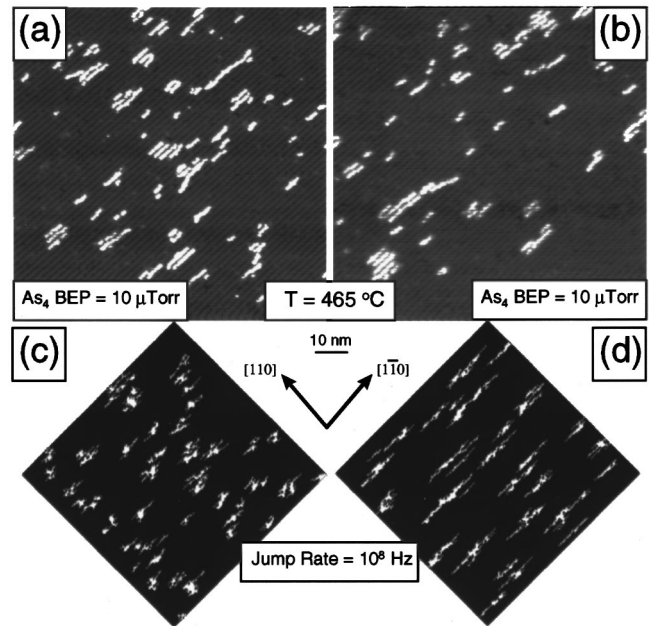


FIG. 5. (a) and (b) Comparison of filled state STM images taken at a sample bias of  $-3.0$  V and a demanded tunneling current of  $0.2$  nA of the  $\text{GaAs}(001)-(2 \times 4)$  reconstructed surface after deposition of 10% of a monolayer of Ga under  $1.0$  and  $10.0 \mu\text{Torr}$   $\text{As}_4$  flux at  $465^\circ\text{C}$ , respectively. (c) and (d) Comparison of simulated STM images from random walk computer simulations. Simulation (c) best fits the  $1.0 \mu\text{Torr}$   $\text{As}_4$  data in (a) using a directional hopping-rate ratio of 100:1 ( $[1\bar{1}0]:[110]$ ). Simulation (d) best fits the  $10.0 \mu\text{Torr}$   $\text{As}_4$  data in (b) using a directional hopping-rate ratio of 5:1 ( $[1\bar{1}0]:[110]$ ).

TABLE I. Comparison of results for different  $As_4$  fluxes.

Quantity	1.0 $\mu$ Torr $As_4$	10.0 $\mu$ Torr $As_4$
directional hopping ( $[1\bar{1}0]:[110]$ )	100:1	5:1
directional sticking ( $[1\bar{1}0]:[110]$ )	100:1	100:1
activation energy (eV)	$1.0 \pm 0.1$	$1.2 \pm 0.1$

5(d) is not visually satisfying, our goal was to change only one parameter, if possible, in the simulation to develop an understanding of the dominant effect of arsenic on the diffusion. Changing only the hop ratio by a factor of 20 provided the best fit to both the island number density and island geometry as a function of substrate temperature. Even though the fit to the aspect ratio is not as good as obtained with the lower arsenic pressure, this is still a much better fit than one could obtain by only changing the stick ratio. Similar to the lower  $As_4$  pressure data, a diffusion coefficient was extracted after combining the Monte Carlo results with the measured island number density. A summary of the parameters used in the simulations for all hopping rates, and the resulting diffusion parameters are displayed in Table I.

#### IV. DISCUSSION

Our Monte Carlo simulations are oversimplified and do not attempt to accurately model the complex processes involved in MBE growth. The input parameters such as the directional hopping-rate and sticking-probability ratios should not be taken literally. These ratios to some extent represent reduced parameters that take into account all processes necessary for growth. For example, the GaAs surface has a  $(2 \times 4)$  reconstruction which is not explicitly modeled, and is therefore folded into the directional hopping-rate ratio. The simulation predicts that the diffusion is faster along the  $[1\bar{1}0]$  direction, which we believe is correct, however, the quantitative prediction of 100 times could easily be in error by a factor of 10. These same comments apply to the directional hopping-rate ratio for the higher  $As_4$  pressure. Even though the error in the directional hopping-rate ratio may be large, we believe the factor of  $\sim 20$  change between the two  $As_4$  pressures is real, since this factor is the only change between the simulations. These same comments apply to the directional sticking-probability ratios. The predicted activation energy is not very sensitive to the directional hopping-rate and sticking-probability ratios, and we believe it is more accurate. However, the energy does represent a weighted average favoring the lowest of the  $[110]$  and  $[1\bar{1}0]$  directional energy barriers.

The highly anisotropic directional hopping-rate ratio for the low  $As_4$  pressure data indicate that the Ga atoms essentially move in one dimension. Once the amount of arsenic was increased by a factor of 10, the island number density dropped by a factor of  $\sim 2$ , indicating that the Ga atoms can now interrogate a larger area. This increase in interrogation area is explained by the factor of  $\sim 20$  decrease in the anisotropy of the directional hopping-rate ratio. This implies that the Ga atom is transformed from a one-dimensional ran-

dom walker to a two-dimensional random walker, which can interrogate more area thus forming a smaller number of islands. We believe the mechanism for this change is due to the excess arsenic lowering the energy barrier to hop in the  $[110]$  direction to a value closer to the barrier height in the  $[1\bar{1}0]$  direction. This is supported by the relatively small change in the activation energy with the factor of ten increase in the arsenic pressure.

Studies of InAs growth on GaAs observed a similar phenomenon; increasing the arsenic pressure decreased the number density of InAs islands.<sup>26</sup> Madhukar *et al.* proposed that the increase in arsenic pressure increased the incorporation rate into existing islands. In terms of our model, this proposal amounts to increasing the sticking probability. In our homoepitaxy study, we have tested the role of the stick ratio, but found that this primarily effects the geometry of the islands not the number density. The most important parameter for effecting the island number density is the hop ratio, which suggests the arsenic effects the mobility of the Ga atoms while diffusing more than directly influencing their incorporation into existing islands. Note, the correct picture is most likely a combination of the two parameters, however, the accuracy of the simulation does not merit such a refinement.

The activation energy for single Ga atom diffusion on the GaAs(001)- $(2 \times 4)$  reconstructed surface has recently been computed using a total-energy first-principles approach.<sup>11</sup> This study determined that the activation energy for diffusion along the  $[1\bar{1}0]$  is 1.2 eV, while diffusion along the  $[110]$  is a much higher 1.5 eV. This difference results in Ga atoms being 100 times more likely to diffuse along the  $[1\bar{1}0]$  direction than along the  $[110]$  direction at normal growth temperatures. This is in good agreement with our low  $As_4$  pressure Monte Carlo predictions. This theoretical prediction is for an ideal defect free surface without any arsenic pressure. Not only is there good agreement with the directional diffusion rates, but also the quantitative value of the activation barrier is in agreement with our value. Of course, our experiment primarily measures the value of the lower barrier. We believe the presence of arsenic effectively lowers the barrier in the  $[110]$  direction to a value closer to 1.2 eV, the result of which is to leave the measured barrier height essentially unchanged, yet enhance the interrogation area.

As mentioned in Sec. I, a large body of literature exists in measuring the properties of MBE growth. A majority of this effort has been spent measuring the diffusion length of the Ga using RHEED oscillations.<sup>3,4,8</sup> The methods described in this study compliment this earlier body of literature. The diffusion length as a function substrate temperature and arsenic flux are visually displayed in the STM images presented in Figs. 1 and 5, as the average distance between islands. It ranges from about 2 to 20 nm.

#### V. CONCLUSION

With the combination of Monte Carlo simulations and *in situ* combined MBE–STM experiments, we were able to measure the activation barrier height, directional hopping-

rate ratio, and directional sticking-probability ratio over a range of sample temperatures and As<sub>4</sub> fluxes. Using STM, the influence of defects can be taken into account, which allows a clearer picture of the basic processes involved in growth. Surprisingly, the Ga atoms on the GaAs(001)-(2×4) reconstructed surface have a higher diffusion rate in a higher As<sub>4</sub> pressure. An activation barrier of ~1 eV is in good agreement with first-principles theoretical predictions. These diffusion parameters may help in modeling the MBE process.

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