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Absence of a step-edge barrier on a polar semiconductor surface with reconstruction

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Based on purely geometrical considerations, we point out that a polar semiconductor surface with reconstruction does not satisfy the necessary condition for the existence of an Ehrlich-Schwoebel (ES) step-edge barrier. We further show by the explicit Monte Carlo calculations that the kinetic surface roughening observed on a GaAs(001) surface can be accounted for by the stability and complexity of the (2×4) reconstruction, and hence an ES barrier is unnecessary to account for it. Finally, we point out that it is incorrect to use the solid-on-solid (SOS) model with an ES barrier for surface growth studies or for atomistic growth simulations.

I. INTRODUCTION

In developing optoelectronic devices, it is highly required to prepare a sample substrate with a very flat and smooth surface. This is especially true for the development of a quantum well in which two-dimensional (2D) electron gas states are realized.[1,2] Accordingly, the control of a kinetic surface roughening in epitaxial growth is important from the viewpoint of both science and engineering. In surface growth kinetics, the relevance of a step-edge barrier, which is often called an Ehrlich-Schwoebel (ES) barrier,[3,4] to kinetic surface roughening and mound formation is often emphasized for both metal and semiconductor surfaces.[5,6] In the presence of an ES barrier, a diffusive adatom is supposed to stick to a step edge from an ascending side, or it is reflected when it approaches a step edge from a descending side. Because of this asymmetry, an ES barrier has been said to give rise to mound formation or kinetic surface roughening, for which extensive computer simulations have been made by assuming *a priori* the presence of an ES barrier.[7–15]

However, since surface atomic structures and electronic properties differ from species to species, it is not at all clear whether an ES barrier exists on a surface in a ubiquitous fashion. Rather, its appearance may strongly depend on some particular surface properties. The purpose of this paper is to give a general argument to clarify the necessary condition for the presence of an ES barrier. Furthermore, we show by the explicit kinetic Monte Carlo (MC) simulations that an ES barrier is unnecessary for the occurrence of kinetic surface roughening or mound formation on a semiconductor surface. This is particularly true for a III-V compound polar semiconductor surface because it is unstable against reconstruction.

This paper is organized as follows. We point out the relationship between the diffusion anisotropy and the presence of an ES barrier in Sec. II. In Sec. III we show by the explicit kinetic MC simulations that, even without an ES barrier, kinetic surface roughening occurs if a surface exhibits nontrivial surface reconstruction. This result calls into question the validity of other simulation works. They are reexamined in Sec. IV where we point out, among other things, that the solid-on-solid (SOS) model and an ES barrier are incompatible to each other. We further point out that even the definition of a kinetic barrier often used for the simulations based on the SOS model with an ES barrier is incorrect. The limitation on the use of the SOS model is also discussed. Sec. V is devoted to a summary.

II. DIFFUSION ANISOTROPY AND A STEP-EDGE BARRIER

We first point out that when the possible role of an atomic step in mound formation was considered by Schwoebel,[4] it was certainly assumed that a step edge should separate two ideally flat and perfectly smooth terraces, on which no effects of surface reconstructions were taken into account. This is because the first real-space observation of the surface atomic structure[16] and the atomic steps[17] were reported more than a decade after his study on the role of a step edge. On an ideally flat and smooth terrace such as those Schwoebel considered, an adatom diffuses and either sticks to a step edge or collides with one or more adatoms to make a cluster, which may still be mobile on a very smooth terrace. This condition of an ideal surface is approximately satisfied on some of the stable metal surfaces, on which large mobile clusters as well as an empty zone in the spatial distribution of mobile atoms near a descending step edge have been observed by using field ion microscopy by Ehrlich and co-workers.[18–23]

It is important to note here that the condition of this smoothness is satisfied approximately on a stable metal surface because of the presence of a delocalized surface electronic state on it, whereas it is not generally satisfied on a semiconductor surface. Moreover, when a surface is not so smooth, growth occurs by the sticking of atoms not only to a step edge but also to defects or other surface sites originating from less-smooth surface atomic structures.[24,25] This property is enhanced when a surface electronic state is highly localized so that the diffusion of an adatom takes place by breaking bonds to atoms in the substrate to leave a dangling bond there and by rebonding with other atoms in the adjacent sites. This hopping diffusion is often seen on a semiconductor surface,[26–29] which usually adopts complicated atomic structures via reconstruction. Thus, a step edge is no longer a special site for an adatom to stick to. This means that an ES barrier may play some roles on a stable metal surface, but not generally on a semiconductor surface.

This consideration is quite general, so that it applies to

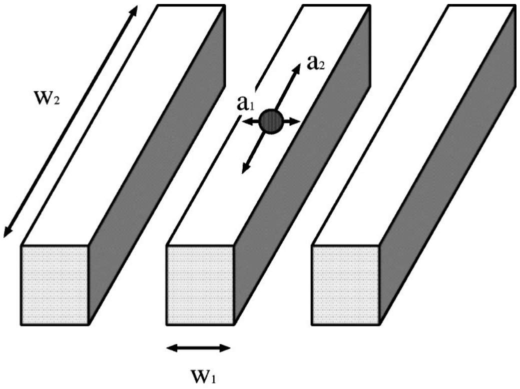

FIG. 1. A trench-and-hill structure, in which the diffusion of an adatom is geometrically restricted. The two perpendicular directions of diffusion for an adatom are indicated by a_1 and a_2, and the corresponding lengths of paths for diffusion on the same atomic layer from one step edge to another are indicated by w_1 and w_2, respectively.

any metal or semiconductor surfaces, irrespective of the presence or absence of an anisotropy in the diffusion of adatoms. For example, diffusion of surface atoms is isotropic on a Si(111) surface, which is known to reconstruct into the very complicated dimer adatom stacking-fault (DAS) structure.[30] Since this structure is not at all atomically flat, it is naturally expected that an ES barrier plays, even if it existed, little role on it. This is supported by the scanning tunneling microscope (STM) observation that the complexity of the DAS structure is sufficient to cause surface roughening.[31–33]

If the diffusion anisotropy exists, we can further proceed to make our argument more concrete. Namely, if an adatom diffuses anisotropically, we must also specify a path of the diffusion so as to define a step-edge barrier associated with it. Let us first consider a case in which this anisotropy is caused by a geometrical restriction, e.g., by the trench-and-hill structure which is often observed on (001) surfaces of III-V materials. The trench-and-hill structure is schematically depicted in Fig. 1, for which we define the two perpendicular directions of diffusion. Correspondingly, we denote the lattice constants in these directions by a_1 and a_2. In these directions, moreover, we define the lengths of paths for an adatom to diffuse on the same atomic layer from one step edge to another by w_1 and w_2, respectively. For definiteness, we assume the inequality $w_1 \leqslant w_2$ to hold, as they are indicated in Fig. 1. Furthermore, we impose that the orientations of these two perpendicular axes a_1 and a_2 are determined such that w_1 takes the smallest possible value. Then, in order for a step edge barrier to be defined, there must exist atomic sites adjacent to and away from an atomic step. For these sites to exist, the inequality $w_1 \geqslant a_1$, or at least the inequality $w_1 > 2a_1$, must be satisfied. Otherwise, no sites away from an atomic step can exist, so that, by its definition, an ES barrier does not exist. In this case, an ES barrier is irrelevant even if it existed along a path in the w_2 direction, since an adatom on top of a hill can easily change its direction of diffusion from the a_2 direction to the a_1 direction, along which an ES barrier is missing.

The typical example of this kind of surface structure is the GaAs(001)-$\beta2(2\times4)$ reconstruction,[34] which is depicted in Fig. 2. In this structure, w_1 is only $2a_1$ and, therefore, the assumption to support the existence of an ES barrier does not hold on it, or on any other similar structures of III-V compound semiconductor surfaces of (001) orientations.

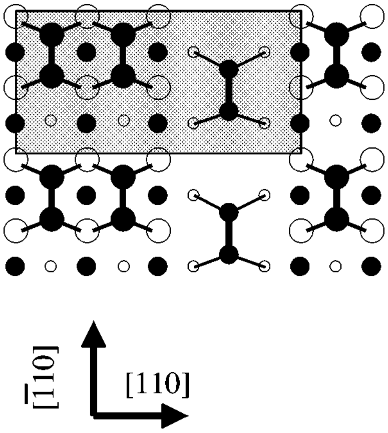

FIG. 2. Plan view of the $\beta2(2\times4)$ structure with the crystallographic directions indicated by the arrows in the lower part. The dark and the bright disks represent As atoms and Ga atoms, respectively, and their radii decrease according to their depths.

Accordingly, when a Ga adatom diffuses along a trench, in which it principally diffuses,[35] to go down a step edge, a barrier hinders this motion since a Ga adatom can no longer find an in-plane Ga atom in the adjacent site to be bonded to. However, this is true not only for the path to go down a step edge onto the lower terrace but also for the path to hop out of a trench to go onto the top of an As-dimer hill of the same terrace. These paths are indicated by the arrows in Fig. 3. Indeed, both the *ab initio*[36] and the molecular-dynamics[37] (MD) calculations revealed that an ES barrier is not associated with the diffusion of a Ga adatom for a descending path down an *A*-step edge, which is parallel to the As-dimer row direction. Moreover, the MD calculation also showed that

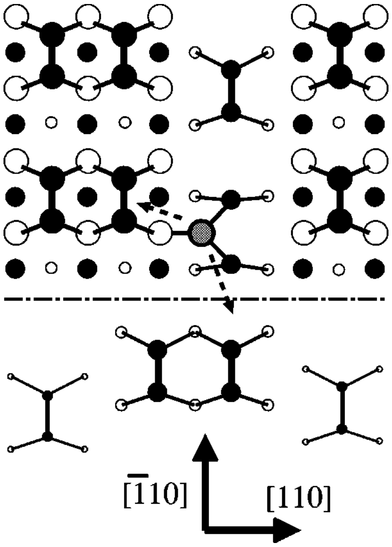

FIG. 3. Two possible diffusion paths for a Ga adatom to go out of a trench site, one to go down a step edge onto the lower terrace, and the other to go onto the top of an As-dimer hill of the same terrace. The horizontal dot-dashed line denotes the border between the upper terrace and the lower one.

the barrier for the diffusion of a Ga adatom near *a straight B-step edge*, which is perpendicular to an As dimer row, is 0.25 eV.[37] As expected, this value is very close to the difference of Ga diffusion barriers between the [110] direction and the [$\bar{1}$10] direction.[36] In other words, the barriers to hop out of a trench site nearly coincides with each other in these calculations irrespective of the difference in the hopping directions.

Here we note that this MD calculation was done in Ref. 37 for a straight *B*-step edge but not for any other shapes of *B* steps, with which short segments of *A* steps are inevitably associated. In other words, for any *B* steps other than a straight one, a Ga adatom can migrate over a descending *B*-step edge via a short segment of an *A*-step edge, so that an ES barrier, even if it existed, plays little role for the diffusion of a Ga adatom on a GaAs(001) surface. Or, equivalently, if this barrier plays any roles for growth, a mound which results from this effect must have a long axis in the [110] direction because the interlayer diffusion in the [$\bar{1}$10] direction is hindered by the presence of an ES barrier. However, the results of the STM observations are opposed to this, i.e., they show that mounds have long axes in the [$\bar{1}$10] direction.[38]

III. KINETIC SURFACE ROUGHENING CAUSED BY SURFACE RECONSTRUCTION

In what follows, we show with an explicit example that kinetic surface roughening does occur on a semiconductor surface even in the absence of an ES barrier if surface reconstruction is properly taken into account in growth studies. Thus, we show that the absence of it does not result in any contradiction with any known phenomena.

To this aim, we carry out atomic-scale growth simulations without introducing any particular effects related to the presence of a step edge, and demonstrate that this barrier is really unnecessary to cause a kinetic surface roughening. To show this, we employ the recently proposed two-species atomic-scale kinetic growth model of GaAs(001) homoepitaxy.[39,40] By using this model, we perform explicit kinetic MC simulations and calculate the kinetic roughness exponent β. For this purpose, we first define the interface width[5,6]

$$w(L,t)=\sqrt{\frac{1}{L^2}\sum_{\mathbf{r}}\,[h(\mathbf{r},t)-\bar{h}(t)]^2}, \tag{1}$$

where L is the linear lattice size, t is the time, $\mathbf{r}$ is the spatial coordinates of surface atoms, and $\bar{h}(t)$ is the average height over the surface at a time t defined by $\bar{h}(t)=(1/L^2)\Sigma_{\mathbf{r}}h(\mathbf{r},t)$. With the use of $w(L,t)$, the exponent β is defined through the relation

$$w(L,t)\sim t^{\beta}. \tag{2}$$

For the growth condition, we use $T=580\,^{\circ}\mathrm{C}$ for the substrate temperature, whereas Ga atomic flux and As$_2$ molecular flux are set to be $f_{\mathrm{Ga}}=0.1$ monolayer per second (ML/s) and $f_{\mathrm{As}_2}=0.4$ ML/s, respectively. With these conditions, we obtained the very large value $\beta\simeq 0.86$ in the range $2.8<\ln t<4.5$, as seen in Fig. 4. During the same interval, this value remains constant for the lattice size ranging from $40a_s\times 40a_s$ to $120a_s\times 120a_s$, as well as for f_{Ga} between 0.05 and 0.20 ML/s when the atomic flux ratio $f_{\mathrm{As}_2}/f_{\mathrm{Ga}}=8$ is held fixed. Here, $a_s=0.40$ nm is the surface lattice constant of GaAs(001).

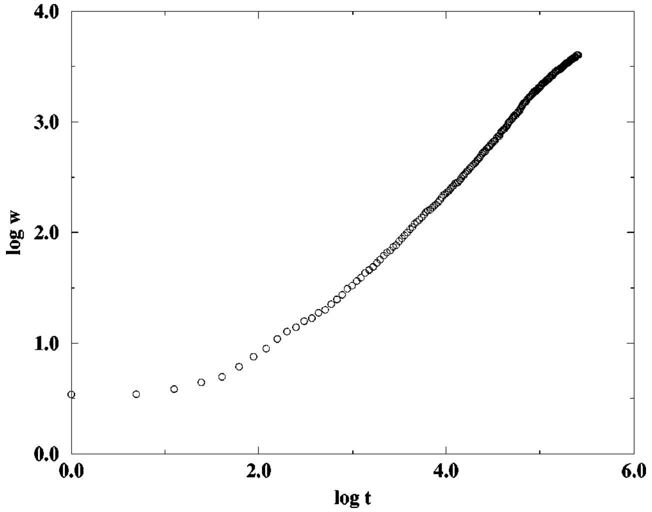

FIG. 4. The log-log plot of the interface width $w(L,t)$ as a function of a time t. The lattice size is $120a_s\times 120a_s$.

Thus, a finite value for the exponent β can be obtained without introducing an ES barrier. The appearance of such a large value of β at the very early stages of a growth is naturally explained by the series of the snapshots in Figs. 5(a)–5(d), showing that a newly generated layer is more favored for Ga adatoms to stick to than older ones. This is well understood by the comparison with Figs. 6(a)–6(d), which show that for a small island just after nucleation to become wider than a single-dimer row, the nearby trenches of the substrate have to be filled. At the same time, the snapshots at a later era in Figs. 6(e)–6(k) show that the As-dimer rows of four-dimer width are rather long lived before they split to

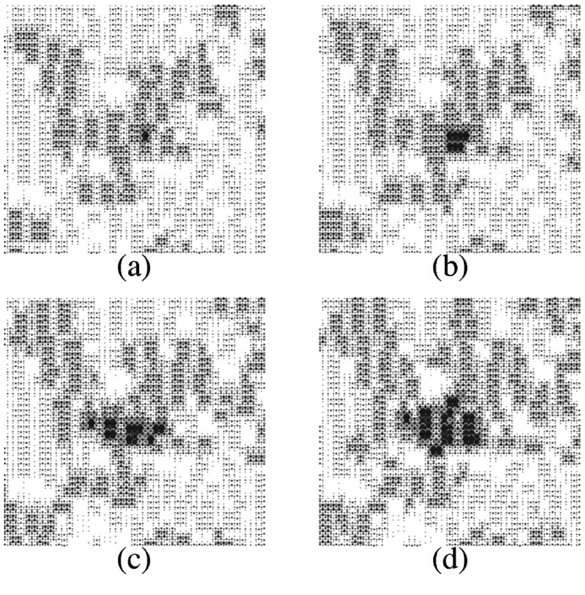

FIG. 5. The series of the snapshots showing the onset of the kinetic surface roughening due to the stability of the $\beta2(2\times4)$ structure. The lattice size used for the calculation is $120a_s\times 120a_s$. $f_{\mathrm{Ga}}=0.1$ ML/s and $f_{\mathrm{As}_2}=0.4$ ML/s are used for the incident fluxes.

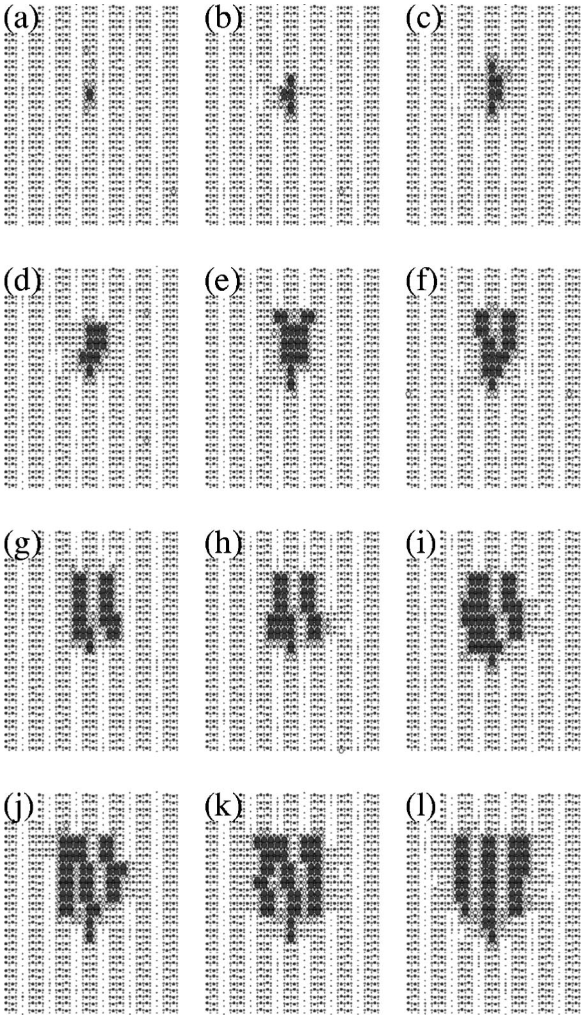

FIG. 6. Snapshots of the nucleation of the $\beta2(2\times4)$ structure on the reconstructed substrate, recorded at every 0.1 s. Dark and bright disks denote As and Ga atoms, respectively. The incident fluxes are $f_{\mathrm{Ga}}=0.1$ ML/s and $f_{\mathrm{As}_2}=0.4$ ML/s.

take the $\beta2(2\times4)$ structure. This means that such a wide island is actually a preferential site for Ga adatoms to stick to, whereby a kinetic surface roughening naturally occurs. Of course the appearance of antiphase boundaries at lower surface layers cooperatively promotes it with their poor reactivity for the sticking of arsenic species. This is true because it is much more difficult to satisfy the geometrical sticking condition of an As_2 molecule than that of a Ga adatom. Not only due to this, but also because Coulomb repulsion works between adjacent As species, they are generally unstable at a phase boundary of the $\beta2(2\times4)$ structure.

Recently, the qualitatively similar experimental results on the *in situ* observations of GaAs(001) homoepitaxy as our simulations were reported by Tanahashi *et al.* by using the scanning electron microscopy.[41] It is worthwhile to note that this phenomenon of kinetic surface roughening and mound formation due to surface reconstruction is already known to occur on a Si(111) surface.[31–33] In addition, in our calculations, this phenomenon is much more promoted than observed because local relaxations of atomic structures are not taken into account in our rigid lattice model. Thus, a newly created layer is more reactive to adatoms for sticking in the model simulations than on a real surface, in exactly the same way as it occurred in the previous rigid lattice model simulations of Si(111) homoepitaxy.[42] Therefore, the estimate $\beta=0.86$ should be regarded as an upper bound of the exponent β in the growth condition used. At the same time, we point out that the values of the roughness exponents strongly depend on growth conditions, because surface reconstructions give additional temporal as well as spatial scales, so that the simple scaling argument cannot be applied to semiconductor surfaces when nontrivial reconstruction is involved in a significant way. This is clearly seen in the recent STM observations, in which it was revealed that the values of the roughness exponents measured on GaAs(001) and InP(001) depend on the III-V flux ratio.[38]

By now, we have argued and explicitly demonstrated the irrelevance of an ES barrier to the occurrence of kinetic surface roughening on a semiconductor surface with reconstruction. Even the shape anisotropy of mounds such as those observed in GaAs(001) homoepitaxy[9] may be well accounted for by energetics due to facet reconstruction.[43–46]

A similar argument can be applied to conclude that the necessary condition for the existence of an ES barrier is not satisfied for the GaAs(111)-$(2\times2)A$ structure or the GaAs(111)-$(2\times2)B$ structure, despite that a Ga adatom diffuses isotropically on them. This is because these surface structures consist of periodic Ga vacancies or As trimers,[47] so that again there are no sites away from a step edge there. Thus, in general, the necessary condition for the presence of an ES barrier is not satisfied on a polar semiconductor surface when it reconstructs to adopt a regular stepped structure so as not to charge it up by making a topmost surface layer composed only of one species.

IV. REEXAMINING OTHER WORKS

In contrast to our argument, some people have performed simulation studies by assuming the relevance of an ES barrier to the mound formation and the kinetic surface roughening on a GaAs(001) surface[9,10] as well as on an InAs(001) surface,[13] the slow recovery of a specular beam reflection high-energy electron diffraction (RHEED) intensity after interrupting growth,[8] and the reentrant layer-by-layer etching of a GaAs(001) surface when AsBr_3 is used as an etchant.[11] However, the kinetic MC calculations in these studies are all based on the one-species SOS model, which is defined not on the realistic zinc-blende structure but on the simple cubic lattice, so that it is not at all clear if such studies are consistent or not.

To examine this, we derive the range of applicability of the SOS model, in which the activation barrier is defined by

$$E=E_s+nE_N, \tag{3}$$

where E_s is the contribution to the barrier from a substrate, and n is the number of nearest neighbors (NN) on a surface, from which an additional contribution is given proportional to E_N.

Here, we pay attention to the fact that the characteristic time scale for surface diffusion τ is given in terms of E_s as $\tau=\nu^{-1}\exp(E_s/k_BT)$, with ν the attempt frequency, k_B the Boltzmann's constant, and T the substrate temperature. As for ν, $\nu=2k_BT/h$ is used,[48] where h is the Planck's constant.

Next, let us suppose that the model has the spatial resolution $R_s = \xi a_s$. If $\xi > 1$, the time resolution of the model is reduced from τ to $\tau_{SOS} = \xi^2 \tau$ on a 2D system. In addition to this, let us denote the deposition rate of Ga atoms by f_{Ga} monolayer per second (ML/s). When a specular RHEED intensity oscillation is observed in the steady-state growth mode, the time resolution for this observation τ_{RHEED} must be an order of magnitude smaller than $1/f_{\mathrm{Ga}}$, i.e., $\tau_{RHEED} \simeq 0.1/f_{\mathrm{Ga}}$. This is because a Ga flux is known to control the growth rate of a GaAs(001) surface in a sufficiently large arsenic overpressure, under which a steady-state growth mode is realized.

Accordingly, as long as τ_{RHEED} is larger than τ_{SOS}, one can compare the oscillations between the SOS model calculations and the specular RHEED intensities. If we substitute $f_{\mathrm{Ga}} = 0.47$ ML/s, $E_s = 1.58$ eV, and $T \geqslant 540\,°\mathrm{C}$, which were used in Refs. 48,49, into the above relations, we can obtain the estimates of ξ. For definiteness, we derive the maximum value of ξ which satisfies the inequality $\tau_{RHEED} \geqslant \tau_{SOS}$, as defined by

$$\xi_m = \sqrt{\frac{\tau_{RHEED}}{\tau}} \simeq \sqrt{\frac{0.1}{\tau f_{\mathrm{Ga}}}}. \tag{4}$$

If we substitute $E_s = 1.58$ eV into the definition of τ, we obtain $\tau = 1.83 \times 10^{-4}$ s at $T = 540\,°\mathrm{C}$ and $\tau = 6.06 \times 10^{-5}$ s at $T = 580\,°\mathrm{C}$, respectively. Therefore, we obtain $\xi_m \simeq 34$ at $T = 540\,°\mathrm{C}$ and $\xi_m \simeq 59$ at $T = 580\,°\mathrm{C}$, respectively. If we further substitute these values and $a_s = 0.40$ nm into $R_s = \xi a_s$, we obtain $R_s \simeq 13.6$ nm at $T = 540\,°\mathrm{C}$ and $R_s \simeq 23.7$ nm at $T = 580\,°\mathrm{C}$. Even when the flux f_{Ga} is increased, ξ_m remains very large, e.g., at $T = 540\,°\mathrm{C}$, $\xi_m \simeq 23$ for $f_{\mathrm{Ga}} = 1.0$ ML/s, and $\xi_m \simeq 10$ for $f_{\mathrm{Ga}} = 5.0$ ML/s, which correspond to $R_s \simeq 9.3$ nm and $R_s \simeq 4.2$ nm, respectively. These results show that such coarse resolutions are sufficient to simulate a steady-state island growth mode. Hence, in spite of the fact that a steady-state island growth mode was shown to be well simulated by the use of the SOS model,[48] this does not guarantee that the SOS model has an atomic-scale spatial resolution.

This further means that the good agreement was obtained in Ref. 48 because all the orientation dependencies of surface growth kinetics are averaged out in this mode to leave a macroscopically relevant quantity only. To see this, let us recall that $E_s = 1.58 \pm 0.02$ eV was obtained on vicinal surfaces by comparing the simulated and observed transition temperatures T_t between a step flow growth mode at $T > T_t$ and an island growth mode at $T < T_t$.[48,50] More precisely, T_t is the temperature where an oscillation of a specular RHEED intensity disappears. Based on the step density model,[51] this phenomenon has been interpreted to occur when Ga diffusion is sufficiently enhanced by the increase of the substrate temperature, so that Ga adatoms are enabled to reach a step edge by diffusing across a terrace. In other words, T_t is the temperature where an island growth mode ceases when the substrate temperature is increased from below. Therefore, this transition is a macroscopic phenomenon. Accordingly, an atomic scale accuracy is not required for the model to fulfill this comparison, and thus, $E_s = 1.58 \pm 0.02$ eV is valid only at a macroscopic scale in exactly the same manner as the diffusion coefficient, $D = a_s^2/\tau$, is a macroscopic quantity. This is consistent with the fact that $E_s = 1.58 \pm 0.02$ eV is substantially larger than the ''bare'' value $E_D = 1.3 \pm 0.1$ eV obtained as the activation barrier for surface Ga diffusion.[51] In other words, E_s is a renormalized quantity,[52] and hence, to use E_s is equivalent to consider an ''effective-field'' theory. In Ref. 8, however, an ES barrier was introduced into the SOS model so as to obtain a good agreement not only for the steady-state island growth mode but also for the recovery process of a specular RHEED intensity after terminating growth. In this study, $E_s = 1.54$ eV was used, i.e., a different value than $E_s = 1.58 \pm 0.02$ eV,[48] but still significantly larger than $E_D = 1.3 \pm 0.1$ eV.[51] However, since an ES barrier is associated with the presence of an atomic step, which is a structure literally at an atomic scale, the introduction of this effect requires a model to possess atomic scale resolution. Therefore, it is inconsistent to use the renormalized value E_s for such simulations together with the use of an ES barrier. This inconsistency in trying to achieve the agreement between simulations and RHEED observations further indicates that the SOS model is unable to simulate kinetic processes of GaAs(001) at an atomic scale.

Even independently of any simulations, we can point out the irrelevance of an ES barrier to the recovery process after terminating growth of GaAs(001) homoepitaxy. Indeed, if an ES barrier is really relevant to this process by any means, one cannot make a well-ordered flat substrate or buffer layers to prepare for growth experiments. This is because, according to this explanation, to carry out a long period of annealing on buffer layers necessarily results in the formation of three-dimensional structures. Therefore, an ES barrier cannot be relevant to a recovery process on it. In other words, the fact that one can experimentally make flat buffer layers on GaAs(001) already excludes the possibility for the existence of an ES barrier.

It is important to note that, since an atomic scale accuracy was not needed in obtaining $E_s = 1.58 \pm 0.02$ eV,[48] this accuracy was not required either for the calculated step density to probe the disappearance of an oscillation. In other words, this result does not guarantee that a step density is relevant to any atomic scale phenomena observed on GaAs(001). Indeed, we pointed out in our recent study that this is exactly the case for a GaAs(001)-(2×4) surface.[53] Namely, even though the time evolutions of a step density and a specular RHEED intensity can be correlated with each other at a macroscopic scale in the steady-state island growth mode, they do not have any causal relations to each other. Since the step density model[51] has been the basis of the kinetic MC simulations with the use of the SOS model, this result precludes the physical relevance of the simulation studies such as the simulations on the step flow growth mode[49] or the recovery process.[8] The only result which remains valid is the estimate of $E_s = 1.58 \pm 0.02$ eV,[48] because the method which was utilized to estimate it does not depend on the details of the atomic scale kinetics. We can see another example on the failure of the combined use of the SOS model and an ES barrier in the simulations of kinetic surface roughening observed in InAs(001) homoepitaxy.[13] In this study, the simulated interface width w was found to reach the minimum value at the layer thickness of about 5 ML, which is smaller than the experimentally obtained value of 0.6 μm via atomic force microscopy observation by as large as three orders of magnitude[13] if the diffusive species of the model has an atomic-scale bilayer thickness of a real InAs(001) surface.

Now that an ES barrier is shown to be irrelevant to kinetic surface roughening, mound formation, or the recovery of a specular beam RHEED intensity after the interruption of growth, the only remaining experimental support for the existence of an ES barrier on GaAs(001) is the experimental discovery of the reentrant layer-by-layer etching by Kaneko *et al.* with the use of $AsBr_3$ as the etchant.[11] Although simulations were performed in Ref.11 to try to account for this phenomenon by introducing an ES barrier to the SOS model, this explanation is again incorrect. This is because the inclusion of an ES barrier inevitably induces a three-dimensional morphology at low temperatures, as Fig. 2 of Ref. 11 undoubtedly shows by the gradual decrease of the mean negative step densities calculated at $T \leqslant 500\,°\mathrm{C}$. In deep contrast to these simulation results, the mean RHEED intensities of the oscillations obtained by their measurement at $380\,°\mathrm{C} \leqslant T \leqslant 420\,°\mathrm{C}$ in Fig. 1 of Ref. 11, i.e., below the transition temperature of the reentrant layer-by-layer etching, are constants of time, indicating that this is entirely a 2D phenomenon. Even Ritz, Kaneko, and Eberl stated explicitly that, above and below the range of the transition, the RHEED pattern is streaked over the duration of the measurement and that the etching process below 420 °C is reaction-rate limited, while it is supply-rate limited above 450 °C.[54] Therefore, it is clear by its definition that an ES barrier has nothing to do with these phenomena. Also, it is inadequate to use the SOS model to simulate the etching phenomenon because it cannot deal with such chemical reactions due to its crude simplicity.

Therefore, there are actually no true evidences to support the existence of an ES barrier on a GaAs(001) surface or on an InAs(001) surface. Incidentally, an ES barrier was introduced into the SOS model in Refs. 6–15 as

$$\tau^{-1} = \nu \exp\{-[E_s + nE_N + \Delta m \Theta(\Delta m) E_B]/k_B T\}, \quad (5)$$

where $\nu = 2k_BT/h$, and the definitions of n and E_N are the same as those in Eq. (3). In addition to them, E_B denotes a barrier associated with a step edge and Δm is the difference in the coordination numbers of the next-nearest-neighbor (NNN) sites in the plane beneath and above the hopping atom before and after the hop. $\Theta(x)$ is a step function, which becomes 1 for $x>0$ and 0 otherwise.

However, according to the transition-state theory, on which the conventional kinetic theories are based, a kinetic barrier must be given by the energy difference between a local minimum and a nearby saddle point of a potential surface *before* a hop.[55] Only with this definition, an Arrhenius form is used to define a rate of a kinetic event. Although there is an option to use an energy difference between before and after a hop,[56] it is incorrect to apply one method to the NN interactions and the other to the NNN interactions separately, because only the total activation barrier at each configuration must be used in MC calculations.[57] Therefore, no results obtained with the use of this ill-defined barrier can be correct.

Here, we note that an apparent resemblance between simulation results and any experimental data does not guarantee the validity of any assumed effects in simulations if they are not given any proper reasonings to exist, as Das Sarma and co-workers pointed out for the case of an ES barrier by the explicit calculations.[58] At least, one must keep it in mind that it is indispensable to carry out order estimations before applying a model to any physical phenomena so as to examine its validity as well as its range of applicability to study them. However, no such statement can be found in Refs. 6–15 in spite of the fact that, in these works, the simulation results based on the SOS model were directly compared with the results of the experimental observations.

In relation to this, let us further discuss whether the SOS model itself can be used to study any atomic scale phenomena. To answer this question, it is important to note that the SOS model is defined on the simple cubic lattice, which is actually not the crystallographic symmetry of any real materials. The difference in the symmetry of an underlying lattice may not matter if the dynamical scaling theories and the kinetic universality hypothesis are valid as they are discussed, e.g., in Refs. 5,6. This literature, however, does not deal with any effects of surface reconstruction in deriving the scaling exponents. In contrast, both our result in Sec. III and the experimental report by Lengel and co-workers[38] provide the evidence that the hypothesis of kinetic universality class is no longer valid when surface reconstruction is involved in the growth of a surface in the nontrivial manner. Moreover, since the kinetic barrier of the SOS model in Eq. (3) depends only on the coordination number, it is too crude to reflect the important properties of atomic scale interactions such as the strong orientation dependence of the atomic bonding of a semiconductor material or the delocalized nature of the electronic states of a metal surface. Although this model was used to simulate growth of a Si(001) surface,[59] the same argument as we did to estimate ξ on GaAs(001) can be made on Si(001), too, to obtain a similar result. Thus, the SOS model cannot be used to study atomic scale growth phenomena on the surface of a real material whenever a nontrivial reconstruction is involved. On the contrary, if such an attempt is made to compare results obtained by simulations based on the SOS model with atomic scale observations, a discrepancy will certainly appear. Indeed, we can see the example of such a discrepancy in Fig. 3 of Ref. 60, where the fine resolution STM images of Si(001) were directly compared with the snapshots of the SOS model simulations without examining its spatial resolution. It is therefore not a big surprise that the results obtained by using macroscopic equations and by the simulations with the SOS model were found to agree well with each other.[61] This is because neither of them are valid microscopically, as opposed to the purpose of Ref. 61 to provide a bridge between microscopic and macroscopic length and time scales.

V. SUMMARY

We pointed out that a III-V compound polar semiconductor surface with reconstruction does not satisfy the necessary condition for the presence of an Ehrlich-Schwoebel (ES) barrier. Instead, by carrying out a kinetic MC simulation of homoepitaxial growth on GaAs(001), we calculated the kinetic roughness exponent β and obtained $\beta = 0.86$ as its upper bound. In this simulation, we did not introduce any effects specific to a step edge. Thus, the emergence of kinetic surface roughening in homoepitaxial growth of GaAs(001) and other similar surfaces is shown to be accounted for by the stability and complexity of surface reconstruction and the need for a structural transformation from a stable structure to transient ones that a surface must experience during epitaxial growth. This is especially true at the initial stage of growth from a well-ordered surface.

Moreover, we examined the range of applicability of the widely used SOS model to show that, in general, it cannot be used to study an atomic scale phenomenon on a solid surface whenever nontrivial surface reconstruction is involved in it. This naturally calls into question the validity of kinetic MC simulations in which the SOS model and an ES barrier are used in combination to simulate the growth of GaAs(001) or InAs(001). We pointed out that all these works are actually incorrect; even the definition of the kinetic barrier used in these studies is incorrect. Thus, there are indeed no evidences, theoretically or experimentally, to support the existence of an ES barrier on a polar semiconductor surface with reconstruction. On the contrary, there are theoretical studies to show its absence at the *A* step of a GaAs(001) surface.

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[1] L. Esaki and R. Tsu, IBM J. Res. Dev. **14**, 61 (1970).

[2] M. J. Kelly, *Low-Dimensional Semiconductors* (Clarendon Press, Oxford, 1995).

[3] G. Ehrlich and F. G. Hudda, J. Chem. Phys. **44**, 1039 (1966).

[4] R. L. Schwoebel, J. Appl. Phys. **40**, 614 (1968).

[5] A. Pimpinelli and J. Villain, *Physics of Crystal Growth* (Cambridge University Press, Cambridge, 1998).

[6] A.–L. Barabási and H. E. Stanley, *Fractal Concepts in Surface Growth* (Cambridge University Press, Cambridge, 1995).

[7] P. Šmilauer, M. R. Wilby, and D. D. Vvedensky, Phys. Rev. B **47**, 4119 (1993).

[8] P. Šmilauer and D. D. Vvedensky, Phys. Rev. B **48**, 17 603 (1994).

[9] M. D. Johnson, C. Orme, A. W. Hunt, D. Graff, J. Sudijono, L. M. Sander, and B. G. Orr, Phys. Rev. Lett. **72**, 116 (1994).

[10] C. Orme, M. D. Johnson, K. T. Leung, B. G. Orr, P. Šmilauer, and D. D. Vvedensky, J. Cryst. Growth **150**, 128 (1995).

[11] T. Kaneko, P. Šmilauer, B. A. Joyce, T. Kawamura, and D. D. Vvedensky, Phys. Rev. Lett. **74**, 3289 (1995).

[12] M. Rost, P. Šmilauer, and J. Krug, Surf. Sci. **369**, 393 (1996).

[13] M. F. Gyure, J. J. Zinck, C. Ratsch, and D. D. Vvedensky, Phys. Rev. Lett. **81**, 4931 (1998).

[14] P. Tejedor, F. E. Allegretti, P. Šmilauer, and B. A. Joyce, Surf. Sci. **407**, 82 (1998).

[15] P. Tejedor, P. Šmilauer, C. Roberts, and B. A. Joyce, Phys. Rev. B **59**, 2341 (1999).

[16] G. Binnig, H. Röhrer, Ch. Gerber, and E. Weibel, Phys. Rev. Lett. **49**, 57 (1982); **50**, 120 (1983).

[17] R. S. Becker, J. A. Golovchenko, E. G. McRae, and B. S. Swartzentruber, Phys. Rev. Lett. **55**, 2028 (1985).

[18] S. C. Wang and G. Ehrlich, Phys. Rev. Lett. **79**, 4234 (1997).

[19] S. C. Wang, U. Küpick, and G. Ehrlich, Phys. Rev. Lett. **81**, 4923 (1998).

[20] A. Gölzhäuser and G. Ehrlich, Phys. Rev. Lett. **77**, 1334 (1996).

[21] K. Kyuno and G. Ehrlich, Phys. Rev. Lett. **81**, 5592 (1998).

[22] K. Kyuno, A. Gölzhäuser, and G. Ehrlich, Surf. Sci. **397**, 191 (1998).

[23] K. Kyuno and G. Ehrlich, Surf. Sci. **387**, L766 (1997).

[24] D. R. Bowler and C. M. Goringe, Phys. Rev. B **58**, 3937 (1998).

[25] K. Cho and E. Kaxiras, in *Multiscale Modelling of Materials*, edited by V. V. Bulatov, T. D. de la Rubia, R. Phillips, E. Kaxiras, and N. Ghoniem, MRS Symposia Proc. No. 538 (Materials Research Society, Pittsburgh, 1999), p. 341.

[26] T. Yamasaki, T. Uda, and K. Terakura, Phys. Rev. Lett. **76**, 2949 (1996).

[27] J. Nara, T. Sasaki, and T. Ohno, Phys. Rev. Lett. **79**, 4421 (1997).

[28] S. Jeong and A. Oshiyama, Phys. Rev. Lett. **79**, 4425 (1997).

[29] S. Jeong and A. Oshiyama, Phys. Rev. Lett. **81**, 5366 (1998).

[30] K. Takayanagi, Y. Tanishiro, S. Takahashi, and M. Takahashi, Surf. Sci. **164**, 367 (1985); J. Vac. Sci. Technol. A **3**, 1502 (1985).

[31] Y. Shigeta, J. Endoh, and K. Maki, Phys. Rev. B **51**, 2021 (1995); J. Cryst. Growth **166**, 617 (1996).

[32] Y. Shigeta, J. Endoh, H. Fujino, and K. Maki, Surf. Sci. **358**, 414 (1996).

[33] C. J. Lanczycki, R. Kotlyar, E. Fu, Y.-N. Yang, E. D. Williams, and S. Das Sarma, Phys. Rev. B **57**, 13 132 (1998).

[34] For a review of the theoretical studies on the GaAs(001)-(2×4) structures, see C. M. Goringe, L. J. Clark, M. H. Lee, M. C. Payne, I. Štich, J. A. White, M. J. Gillan, and A. P. Sutton, J. Phys. Chem. **101**, 1498 (1997); and for the experimental studies, Q.-K. Xue, T. Hashizume, and T. Sakurai, Prog. Surf. Sci. **56**, 1 (1997).

[35] A. Kley, P. Ruggerone, and M. Scheffler, Phys. Rev. Lett. **79**, 5278 (1997).

[36] A. Kley (unpublished).

[37] M. A. Salmi, M. Alatalo, T. Ala-Nissila, and R. M. Nieminen, Surf. Sci. **425**, 31 (1999).

[38] G. Lengel, R. J. Phaneuf, E. D. Williams, S. Das Sarma, W. Beard, and F. G. Johonson, Phys. Rev. B **60**, R8469 (1999).

[39] M. Itoh, G. R. Bell, A. R. Avery, T. S. Jones, B. A. Joyce, and D. D. Vvedensky, Phys. Rev. Lett. **81**, 633 (1998).

[40] M. Itoh, Ph.D. thesis, University of London, 1999.

[41] K. Tanahashi, Y. Kawamura, N. Inoue, Y. Homma, and J. Osaka, J. Cryst. Growth **188**, 205 (1998).

[42] M. Itoh, Phys. Rev. B **57**, 14 623 (1998).

[43] N. Moll, A. Kley, E. Pehlke, and M. Scheffler, Phys. Rev. B **54**, 8844 (1996).

[44] E. Pehlke, N. Moll, and M. Scheffler, *Proceedings of the 23rd International Conference on the Physics of Semiconductors*, edited by M. Scheffler and R. Zimmermann (World Scientific, Singapore, 1996), pp. 1301–1304.

[45] E. Pehlke, N. Moll, A. Kley, and M. Scheffler, Appl. Phys. A: Mater. Sci. Process. **65**, 525 (1997).

[46] N. Moll, M. Scheffler, and E. Pehlke, Phys. Rev. B **58**, 4566 (1998).

[47] W. Münch, *Semiconductor Surfaces and Interfaces*, Springer Series in Surface Sciences Vol. 26 (Springer, Berlin, 1995).

[48] T. Shitara, D. D. Vvedensky, M. R. Wilby, J. Zhang, J. H. Neave, and B. A. Joyce, Phys. Rev. B **46**, 6815 (1992).

[49] T. Shitara, D. D. Vvedensky, M. R. Wilby, J. Zhang, J. H. Neave,

and B. A. Joyce, Phys. Rev. B **46**, 6825 (1992).

[50] We denote the transition temperature by T_t and not by T_C, as Shitara *et al.* did in Refs. 48,49 since this transition is not a critical phenomenon. This is because the average terrace width of a vicinal surface gives the characteristic length scale associated with this transition.

[51] J. H. Neave, P. J. Dobson, B. A. Joyce, and J. Zhang, Appl. Phys. Lett. **47**, 100 (1985).

[52] J. Tersoff, M. D. Johnson, and B. G. Orr, Phys. Rev. Lett. **78**, 282 (1997).

[53] M. Itoh and T. Ohno (unpublished).

[54] M. Ritz, T. Kaneko, and K. Eberl, Appl. Phys. Lett. **71**, 695 (1997).

[55] S. Glasstone, K. J. Laidler, and H. Eyring, *The Theory of Rate Processes* (McGraw-Hill, New York, 1941).

[56] M. E. J. Newman and G. T. Barkema, *Monte Carlo Methods in Statistical Physics* (Clarendon Press, Oxford, 1999), Chap. 11.

[57] K. Binder and D. W. Heermann, *Monte Carlo Simulation in Statistical Physics, An Introduction* (Springer-Verlag, Berlin, 1998).

[58] S. Das Sarma, P. Punyuindu, and Z. Toroczukai, cond-mat/9908013 (unpublished).

[59] S. Clarke and D. D. Vvedensky, Phys. Rev. Lett. **58**, 2235 (1987); Phys. Rev. B **36**, 9312 (1987); Surf. Sci. **189**, 1033 (1987); J. Appl. Phys. **63**, 2272 (1988); Phys. Rev. B **37**, 6559 (1988).

[60] B. Voigtländer, T. Weber, P. Šmilauer, and D. E. Wolf, Phys. Rev. Lett. **78**, 2164 (1997).

[61] R. E. Caflisch, W. E, M. F. Gyure, B. Merriman, and C. Ratsch, Phys. Rev. E **59**, 6879 (1999).