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## Apparent activation energy during surface evolution by step formation and flow

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## Apparent activation energy during surface evolution by step formation and flow

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**Abstract.** During growth and etching by step flow as examples of anisotropic surface processing the apparent activation energy of the growth/etch rate depends on orientation, increasing gradually as a principal, terrace-rich surface is approached. This behaviour is traditionally explained as a change in the dominating process from step propagation to island/pit nucleation. We show that the orientation dependence of the activation energy is actually the result of a traditionally disregarded temperature dependence in the number of active step sites and is not attributable to an increasing role of step nucleation nor to a purely geometrical decrease in the number of step sites. This modifies the traditional picture of the apparent energy for a principal surface and explains how the energy can be higher than, equal to or even lower than that for vicinal orientations.

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**1. Introduction**

In growth and etching as examples of surface processing, step flow on vicinal surfaces is a typical regime in which the surface evolves by essentially adding/removing atoms at the steps. As the exact orientation of a close-packed principal surface is approached, reducing the number of steps and increasing the number of terraces, the formation of the steps through island/pit nucleation is often perceived as a prerequisite for step flow. Correspondingly, the macroscopic activation energy of the growth/etch rate is considered to reflect this change from step to terrace number dominance and is expected to increase as the principal orientation is approached. This picture of the macroscopic activation energy as faithfully correlated to the microscopic activation energy of the majority sites seems to be in good agreement with most experiments, where typically the apparent energy is largest for the terrace-rich surface. In this paper, however, we present theoretical evidence that the macroscopic activation energy need not be largest for this orientation. By focusing on the particular case of anisotropic etching, we describe how the value of the macroscopic activation energy is obtained from the microscopic activation energies as a non-trivial average and explain how the apparent activation energy can increase, stay constant or even decrease when approaching the principal orientation under step flow conditions. As described below, our findings offer support to previous (somewhat controversial) experimental results where the activation energy was found to be minimum at the terrace-rich orientation.

The central idea of the paper is that surface processing in general and anisotropic etching in particular is a step flow process, itself the result of kink propagation. Thus, the kink sites and not the terrace sites are typically the key protagonists controlling the apparent value of the macroscopic activation energy. This means that the experimentally measured activation energy corresponds essentially to that of the kinks, possibly averaged together with other active step sites. This by itself may not sound completely new to growth experts but it may initially look strikingly odd to etch researchers. A more important issue is that our analysis reveals the presence of an unexpected twist: the observed apparent energy is actually a modified version of the microscopic energy of the kinks according to the dependence of their number on temperature, as described mathematically in the paper. An interesting observation is that the correction term can be positive or negative, leading to an increased or decreased apparent activation energy as the crystal orientation approaches the terrace-rich surface. We show in this way that the activation energy of the principal orientation can be larger than, equal to or even smaller than the activation energy of the vicinal surfaces.

The step flow aspects of anisotropic etching can be traced back to the STM observations by Allongue *et al* [1] and the theoretical exploration of the similarities between etching and growth by Elwenspoek [2]. Much of the experimental microscopy and spectroscopy done for etching in the early and mid-1990s [3]–[5] uses the concept of step propagation. Following these pioneering works for etching, the idea of step flow matured during the late 1990s and atomistic simulators using it in one way or another started to appear [6]–[9]. The first simulations of etching as a step flow process were those from Flidr *et al* using the (kinetic) Monte Carlo method [6].

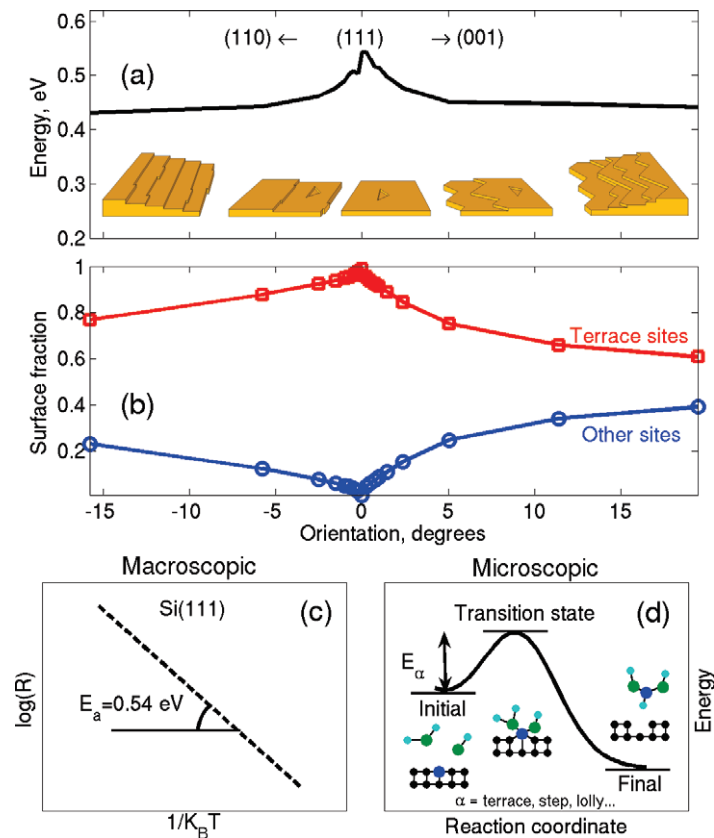
The concept of step flow was developed earlier in growth, starting with the classical paper by Burton *et al* [10], where growth is reduced to the propagation of existing steps, their nucleation (by island formation) and their annihilation (by coalescing of islands and terraces). The central role of the kink sites as the actual locations where growth takes place has been stressed in many reports, e.g. [11]. However, we are not aware of any growth study where the apparent activation energy for a surface has been linked to the activation energy of the kinks and/or other active sites.

In etching, Elwenspoek pointed out the importance of the kinks as one of the weakest sites for the removal of atoms [2]. However, he described the apparent activation energy using classical nucleation theory concluding that the activation energy is essentially the ratio of the square of the step free energy (i.e. the work required to create a step of unit length) to the chemical potential (i.e. the energy cost of removing one atom from the flat surface). According to Seidel *et al* [12], the apparent activation energy corresponds to the energy for breaking the backbonds of the majority sites. None of these descriptions can explain why the activation energy of (111) can be smaller than for vicinal surfaces and other principal orientations, a fact that has been actually measured in a few experiments [13]–[15].

## 2. Macroscopic and microscopic activation energies

Figure 1(a) shows an example of the typical increase in the apparent (or macroscopic) activation energy as the surface orientation approaches (111) during anisotropic etching of crystalline silicon in our simulations. Similar behaviour is observed experimentally [12, 16]. The outlined surfaces in the figure illustrate the fact that the number of steps decreases as the exact orientation is reached. Figure 1(b) shows that the surface fraction of terrace sites increases with improved alignment. The apparent activation energy is defined for each orientation as the slope of the etch rate  $R$  in an Arrhenius diagram (i.e. the slope of the plot  $\log(R)$  versus  $1/T$ , where  $T$  is the temperature in kelvin), as indicated in figure 1(c) for the (111) orientation. The typical correlation between the macroscopic activation energy (figure 1(a)) and the surface fraction of terrace sites (figure 1(b)) has led to the widely used concept that the energy at the exact (111) orientation corresponds to the microscopic energy for the removal of a terrace site atom. The observed decrease from the maximum is regarded as an increasing contribution from the non-terrace sites, whose atomistic activation energies are assumed to be smaller. Here, the microscopic (or atomistic) activation energy refers to the energy difference between an initial state and an activated complex (see figure 1(d)), as considered in standard transition-state theory. The energy barrier ( $E_\alpha$ ) must be overcome in order to proceed with the formation of the reaction products.

Incidentally, the apparent energy has also been reported to decrease as Si(111) is approached, as shown in figure 2 [14, 15]. This behaviour is completely unexpected within the previous picture of the activation energy, and raises questions about its interpretation as a mere reflection of the



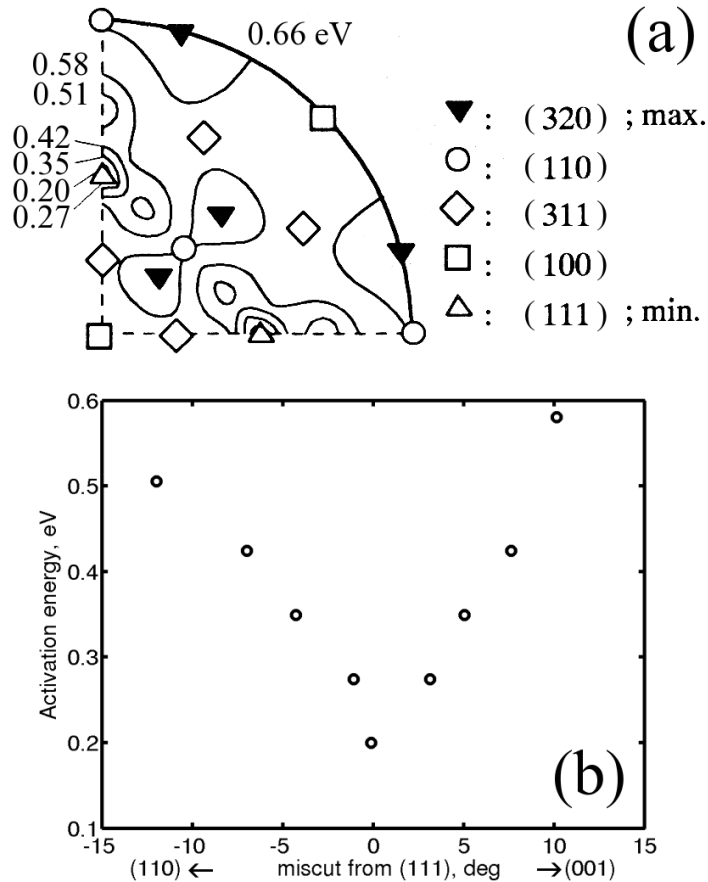
**Figure 1.** (a) Typical dependence of the macroscopic activation energy on surface orientation in the vicinity of Si(111), according to our simulations. (b) Number of terrace sites, normalized to the total number of sites. (c) Definition of the macroscopic activation energy. (d) Simplified picture of the microscopic activation energy.

microscopic energy of the majority sites. More recently, a very careful and accurate experiment by Tan *et al* [13] has shown that the apparent activation energy for (111) is consistently lower than that for (100), questioning again the accepted interpretation and adding controversy. The present study shows how the usual increase and the unexpected decrease in the energy can be both rationalized in terms of the temperature dependence of the number of kinks and other minority sites. Our results completely modify the current picture of the apparent energy by showing that the terrace sites have an insignificant contribution to the macroscopic energy, even when their removal is perceived to be a prerequisite for step formation and flow at the exact principal surface.

### 3. Relation between macroscopic and microscopic activation energies

The apparent activation energy  $E_a$  typically measured experimentally from an Arrhenius plot is the slope of the macroscopic etch rate  $R$  in that plot (figure 1(c)),

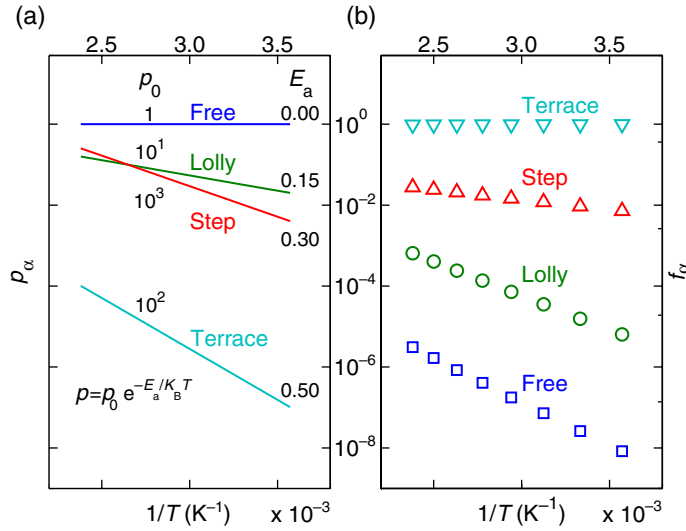
$$E_a = -\partial(\log R)/\partial\beta, \quad (1)$$



**Figure 2.** (a) Contour plot of the macroscopic activation energy as a function orientation for anisotropic etching of crystalline silicon. Etch rates measured using a spherical specimen and a slit mask pattern for vicinal (111) orientations. (b) Orientation dependence of the activation energy along (110)-(111)-(001). 40wt% KOH. Temperature range 40–78°. After [14].

where  $\beta$  is the inverse temperature ( $\beta = 1/k_B T$ ). The etch rate  $R$  is best expressed in terms of the surface fractions ( $f_\alpha = N_\alpha/N$ ) and reaction (removal) rates ( $p_\alpha$ ) [17]:  $R = N \sum_{\alpha=1}^M f_\alpha p_\alpha$ . Here  $M$  is the number of site types (e.g. terrace, step, kink, ...),  $N$  is the total number of surface atoms (distributed between the different sites) and  $\alpha$  is an index denoting the different sites. For instance,  $N_\alpha$  is the number of atoms of type  $\alpha$  and  $p_\alpha$  is the probability (or normalized rate) of removing a particle of that type. A most important feature that is easily overlooked is the fact that the surface fractions  $f_\alpha$  are functions of temperature. This simply results from the fact that the reaction rates themselves depend on temperature (typically according to an Arrhenius behaviour  $p_\alpha = p_{0\alpha} e^{-E_\alpha/K_B T}$  that is different for each surface site) and, correspondingly, the surface will look different at different temperatures. Since both  $p_\alpha$  and  $f_\alpha$  are functions of temperature, simple derivation from equation (1) shows that the macroscopic activation energy  $E_a$  is the sum of two terms [17],

$$E_a = E_p + E_f = \sum_{\alpha=1}^M w_\alpha E_{p_\alpha} + \sum_{\alpha=1}^M w_\alpha E_{f_\alpha}. \quad (2)$$



**Figure 3.** Typical Arrhenius plots for (a) the reaction rates, and (b) the surface fractions.  $E_a$  in eV.

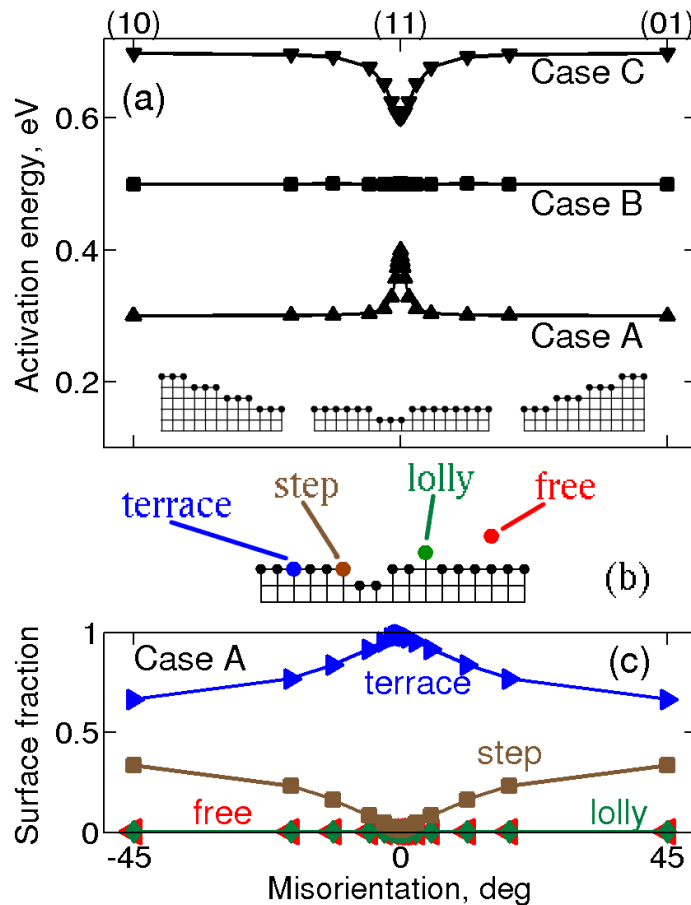
The first term,  $E_p = \sum_{\alpha=1}^M w_{\alpha} E_{p_{\alpha}}$ , is the weighted average over the atomistic activation energies  $E_{p_{\alpha}}$ , where the weight  $w_{\alpha}$  is the *success fraction* for the removal of  $\alpha$ -type sites,  $w_{\alpha} = f_{\alpha} p_{\alpha} / (\sum_{\beta=1}^M f_{\beta} p_{\beta})$ , denoting the number of removed atoms from  $\alpha$ -sites relative to the total number of removed atoms. If one could determine experimentally how many particles of each type are removed (and, thus, the overall number of removed particles) then one could measure the success fractions in an experiment. However, we are not aware of any experimental technique that can perform these measurements. The second term,  $E_f = \sum_{\alpha=1}^M w_{\alpha} E_{f_{\alpha}}$ , involves the *collective* activation energies  $E_{f_{\alpha}}$  associated to the temperature dependence of the surface fractions. In other words,  $E_{f_{\alpha}}$  is the slope of  $f_{\alpha}$  in an Arrhenius plot in just the same manner as  $E_{p_{\alpha}}$  is the slope of  $p_{\alpha}$  (figure 3). Although  $E_{p_{\alpha}}$  corresponds to an energy barrier in transition-state theory (figure 1(d)), an atomistic interpretation of  $E_{f_{\alpha}}$  is not possible since it is only defined for the complete system. By re-writing equation (2) as  $E_a = \sum_{\alpha=1}^M w_{\alpha} (E_{p_{\alpha}} + E_{f_{\alpha}})$ , the fractional contribution of each site to the global activation energy is

$$\epsilon_{\alpha} = \frac{w_{\alpha} (E_{p_{\alpha}} + E_{f_{\alpha}})}{\sum_{\beta=1}^M w_{\beta} (E_{p_{\beta}} + E_{f_{\beta}})}. \quad (3)$$

It is important to notice that the activation energy of equation (2) largely differs from a simple average over the microscopic activation energies, i.e.  $E_a \neq E_a^*$ , where

$$E_a^* = \sum_{\alpha} f_{\alpha} E_{p_{\alpha}}. \quad (4)$$

At first glance, it would seem that the simple average of equation (4) describes the correlation observed between  $E_a$  and  $f_{\text{terrace}}$  (see figures 1(a) and (b)) since it gives a large weight to the majority sites. Actually, it seems that etch researchers widely believe that the activation energy



**Figure 4.** (a) Macroscopic activation energy as a function of misorientation from (11) for three parameter sets. (b) Relative presence of the surface sites (Case A,  $T = 320$  K).

should correspond to equation (4). However, such an average has been shown to be valid only for over-simplified systems (namely, the Thermal Flipping Chessboard) where the transition probabilities between the different sites are independent of temperature [17]. In the real world, the combination of the reaction rates and the existence of an underlying geometry results in site transition probabilities that are inherently temperature-dependent and the correct expression for the activation energy is that of equation (2).

#### 4. Application to a one-dimensional (1D) interface

In order to study which surface site has the leading contribution to the activation energy we consider the 1D interface of a 2D square lattice with variable orientation ( $hk$ ), as depicted in figure 4(a). Typical terrace, step and lolly sites are considered, as shown in figure 4(b). In this system, the ideal crystallographic cut of the (11) orientation presents only terrace sites, the ideal (01) orientation displays only steps, and the crystallographic cut of any intermediate orientation consists of (11) terraces separated by steps. Atoms are assigned Arrhenius removal

rates  $p_\alpha = p_{0\alpha}e^{-E_{p\alpha}/k_B T}$  (cf figure 3(a)), where  $\alpha$  denotes the site type. Occasionally, free atoms may become part of the interface as a result of momentary formation of an overhang and successive backbone destruction.

The values for the removal rate prefactors  $p_{0\alpha}$  and microscopic activation energies  $E_{p\alpha}$  are chosen to mimic step-flow etching conditions, where pit nucleation is a relatively rare event as compared to step flow. Three possible scenarios are considered, depending on the relative value of the microscopic activation energies at the terrace and step sites. This is done by keeping the activation energy constant at the terrace site while considering three different energies for the step site. In Case A we use the values shown in figure 3(a); in Cases B and C we use  $E_a = 0.50$  eV (with  $p_0 = 10^5$ ) and  $E_a = 0.75$  eV (with  $p_0 = 10^8$ ) for the step site, respectively.

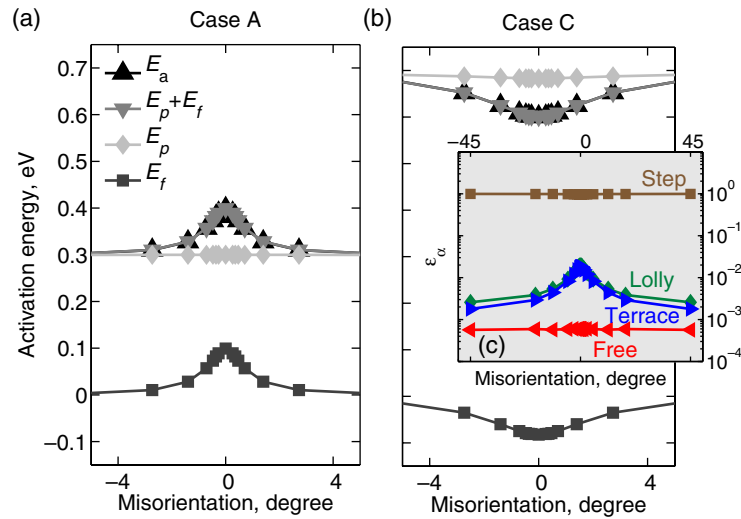
We have also considered more complex systems with an increased number of distinct surface sites, such as the 2D interface between crystalline silicon and the etchant, as sketched in figure 1(a). However, the results of the study are identical and experience has taught that presentation of the results with an increased number of site types is obscured by the larger number of technicalities. Thus, we focus in this paper on the results obtained with the 1D interface.

The macroscopic activation energy obtained from kinetic Monte Carlo simulations of etching in a representative set of misorientations from (11) is shown in figure 4(a) for the three cases A, B and C. This shows that the apparent activation energy may increase (Case A), remain constant (Case B) or decrease (Case C) as the principal (11) orientation is approached. At first glance, since  $E_{p_{\text{terrace}}} = 0.5$  eV in all three cases and  $E_a$  seems to converge towards this value, one may be tempted to conclude that the apparent macroscopic activation energy is governed by the terrace sites according to equation (4). Especially considering the fact that the surface fraction of terrace sites increases as (11) is approached in the three cases, as shown in figure 4(b) for Case A as an example. However, a plot of  $\sum_{\alpha}^M f_{\alpha} E_{p\alpha}$  (not shown) produces completely different curves from those shown in figure 4(a) for Cases A and C, describing correctly only the trivial curve for Case B.

In contrast, figure 5 shows that equation (3) accurately describes the orientation dependence of the apparent activation energy. According to figure 5(a), the increase of  $E_a$  in Case A is completely due to the term  $E_f$ . Only when summing  $E_p + E_f$  one can successfully describe the orientation dependence of  $E_a$ . Similarly, figure 5(b) shows that the decrease of  $E_a$  in Case C is also due to the term  $E_f$  and that the variation of  $E_a$  is only matched after summing the two terms  $E_p + E_f$ . A similar analysis is obtained for Case B.

In order to obtain figure 5, the macroscopic activation energy  $E_a$  is determined for each orientation directly from the slope of the etch rate in an Arrhenius plot after simulating the process at eight different temperatures. Similarly, the collective activation energies  $E_{f\alpha}$  are determined by plotting the surface fractions against the inverse temperature for each orientation, in a manner similar to that presented in figure 3(b) for the case of (11). The two contributions  $E_p = \sum_{\alpha} w_{\alpha} E_{p\alpha}$  and  $E_f = \sum_{\alpha} w_{\alpha} E_{f\alpha}$  to the macroscopic activation energy (equation (2)) are determined by using the input values for  $E_{p\alpha}$ , the obtained values for  $E_{f\alpha}$  and the success fractions  $w_{\alpha}$  (averaged over the eight temperatures). Figures 5(a) and (b) show that without the term  $E_f$  it would be impossible to describe the macroscopic activation energy  $E_a$  satisfactorily using only the term  $E_p$ , stressing the fact that the temperature dependence of the surface fractions plays a crucial role.

Since equation (2) explains satisfactorily the overall variation of the macroscopic activation energy, we proceed further and determine the relative contribution of each site to  $E_a$  by using equation (3). This is done in figure 5(c) for Case C. The figure demonstrates that, although the contribution from the terrace sites becomes more important as (11) is approached, the



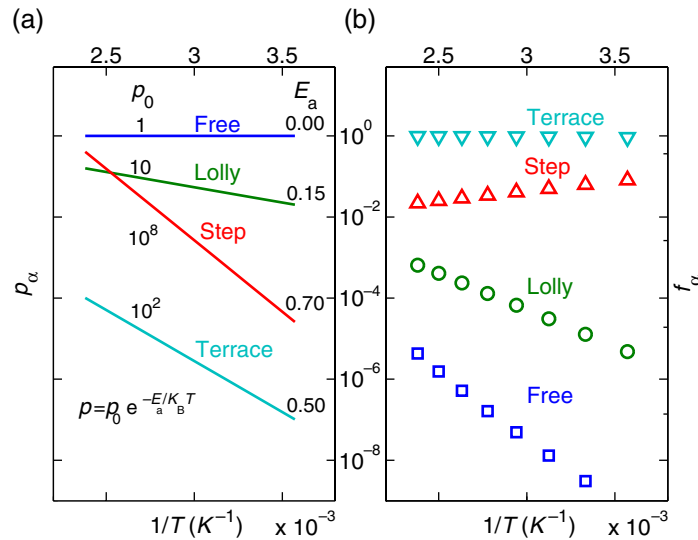
**Figure 5.** The apparent activation energy as the sum of two terms for (a) Case A and (b) Case C. (c) Relative contribution of each site to the apparent activation energy (for Case C).

participation is only about 2% at most while the step sites contribute more than 95%. Similar results are obtained for Cases A and B (not shown).

## 5. Discussion

By combining figures 5(b) and (c), we conclude that the activation energy is governed by the step sites and, in particular, by the temperature dependence of their surface fractions. Figure 5(c) shows that the step sites have by far the leading contribution to the macroscopic activation energy and figure 5(b) shows that it is the temperature dependence of the surface fractions (materialized in the form of  $E_f$ ) that explains the orientation dependence of  $E_a$ . It is this term that completely determines whether the activation energy will reach a minimum, a maximum or will remain constant. In this respect, when the surface fraction of step sites increases with temperature, the macroscopic energy goes through a maximum; and when it decreases, the energy experiences a minimum. As an example, figure 3(b) shows that the surface fraction of steps increases with temperature for Case A while figure 6(b) shows that it decreases for Case C. These figures, which correspond to the (11) orientation, are similar for all other vicinal orientations. For Case B, the surface fraction of step sites remains constant with temperature and the macroscopic energy is directly given by the atomistic activation energy at the step sites ( $E_{p_{\text{step}}} = 0.5$  eV).

From a theoretical perspective, it would be of interest to know whether the step sites can have smaller or larger activation energy than the terrace sites. If we assume that the transition state is the same for all sites, the difference in their activation energies originates from the energy difference between the sites themselves. Since usually this energy correlates with the number of bonds, it is traditionally accepted that the activation energy for the terrace should be larger than for the step. However, deviations from this picture may occur due to additional phenomena, such as e.g. micromasking by metal impurities, which might preferentially adsorb at the step sites. We are currently performing *ab initio* studies in order to understand the details



**Figure 6.** Arrhenius plots for (a) the reaction rates and (b) the surface fractions for orientation (111) (Case C).  $E_a$  in eV.

of the interaction of several impurities with the H-terminated and/or OH-terminated silicon step sites. The temperature behaviour of the stability of these micromasks will affect the number of step sites, making it increase or decrease with temperature. As shown in the present study, this will determine whether the macroscopic activation energy will increase or decrease when approaching the principal orientation.

Recently, a careful experiment by Tan *et al* [13] has shown that the apparent activation energy for (111) is consistently lower than that for (100) if widely separated V-grooves are used for determining the etch rate but it is larger if the mask involves smaller distances between the patterns (e.g. by using a wagon wheel mask), which suggests that diffusion phenomena have a role. Diffusion refers to the transport of the reactants and/or products to/from the locations where they are consumed/produced, which typically takes a longer time than the atomistic reactions. This time delay results into the formation of etchant depletion regions, thus affecting the etch rate. Even though the experimental results in [13] concern 35wt% KOH only, similar results are expected for other concentrations.

From our perspective, these experimental results suggest that the surface fraction of kink sites can decrease with increasing temperature due to diffusion. In normal conditions, raising the temperature will typically result in a larger number of removed particles for all kinds and, although additional kinks are removed, more kinks are created due to the removal of other types. Thus, there are more kinks at high temperature than at low temperature. This is the typical case resulting into a higher apparent activation energy for (111). However, diffusion transport can modify this situation. Due to the higher etch rates at higher temperature, more etchant is consumed and etchant depletion may develop, as suggested in [13]. This can slow down the production of kinks without significantly affecting their removal rate. As a result, we end up with fewer kinks on the surface at higher temperature and a lower apparent activation energy for (111).

The three cases analysed in the present study provide a simple manner of controlling the temperature dependence of the number of steps. In an experiment, external factors

such as micromasking and/or diffusion may be at the root of the temperature dependence of this number.

## 6. Conclusions

In conclusion, during typical surface processing conditions leading to step formation and flow, the majority sites have an insignificant contribution to the activation energy. As a result, the traditional picture of the apparent energy for a principal surface as the activation energy for island/pit nucleation must be abandoned. The usual increase in the macroscopic activation energy as the principal orientation is approached is due to the temperature dependence of the surface fractions of the minority sites. Depending on the details of this temperature dependence the apparent energy may experience a maximum, be constant or cross a minimum at the principal orientation.

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