Monolayer oxidation on Si(001)-(2×1) studied by means of reflectance difference spectroscopy

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Thermal growth of the first oxide layer on a single-domain Si(001)-(2×1) surface has been investigated by means of reflectance difference spectroscopy. The transition temperature between two different growth modes, namely, Langmuir-type adsorption and two-dimensional island growth, was identified from Arrhenius plots of the oxidation periods for the growth of an oxide monolayer. The activation energy for the growth of an oxide monolayer was estimated to be $\epsilon_1 = 0.16 \pm 0.03$ eV. The activation energy at low coverage was estimated to be $\epsilon_1^* = 0.26 \pm 0.03$ eV from analysis of the initial slope of the uptake curves. Our results demonstrate that a finite activation energy exists for monolayer oxide formation on Si(001)-(2×1) at high temperatures.

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I. INTRODUCTION

Surface-sensitive optical spectroscopies, such as reflectance difference spectroscopy (RDS) and surface differential reflectance (SDR) spectroscopy, have attracted considerable interest because of their wide range of applicability to both semiconductor and metal surfaces.[1] These methods provide us with spectral fingerprints associated with particular surface structures, and real-time measurements of the spectral intensity may reveal various chemical reactions proceeding simultaneously. The study of oxidation on silicon surfaces provides a benchmark for these techniques since there are several oxygen adsorption sites, leading to structural complexity of the oxide islands.[2]

Thermally grown SiO_2 film with a thickness of about 1 nm is of vital importance since it can be used as a buffer layer for the growth of high-*k* materials.[3] An oxide thickness of 1 nm corresponds to three or four oxide layers. From a technological point of view, the growth of each of these layers is equally important. It was pointed out that the growth of an oxide monolayer on Si(001) has a significant influence on the growth kinetics of the second layer of the oxide.[4] Thus, the growth kinetics at the initial stage of oxidation, especially monolayer formation, is of great interest. The initial growth modes have recently been identified by real-time measurement with Auger electron spectroscopy (AES),[5] ultraviolet photoemission spectroscopy (UPS),[6] and x-ray photoemission spectroscopy.[7]

Oxidation modes on Si(001) can be classified into two regimes. Over a wide range of pressure-temperature conditions, there is a critical curve which separates the oxidation modes for which etching is the dominant process (active oxidation regime) from those which produce SiO_2 growth (passive oxidation regime).[8] The passive oxidation modes have been recently identified by real-time measurement with photoemission spectroscopic methods.[5–7] The passive oxidation can be subdivided into Langmuir-type adsorption and the two-dimensional island growth.[5,6] At an oxygen pressure of 2.7×10^{-5} Pa and a temperature between 853 and 913 K, the uptake curve of O(2*p*) intensity exhibits an exponential curve for Langmuir-type adsorption, while it follows a sigmoid curve for two-dimensional island growth.[6] This transition of the monolayer regime has been further studied with AES both at 2.7×10^{-5} and at 3.7×10^{-4} Pa.[6] Our recent study at the relatively low pressure of 1.0×10^{-5} Pa with real-time SDR showed that the two growth modes, Langmuir-type adsorption and two-dimensional island growth, can be distinguished from the uptake curves for monolayer oxidation with this technique, as with other spectroscopic methods.[9] This result confirmed the feasibility of using such an optical method to estimate the coverage of adsorbates with the same precision as AES.

Here, we describe the growth kinetics of an oxide monolayer on Si(001) as evaluated with RDS, which provides information complementary to that obtained with SDR. It is recognized that SDR excels at precise measurement of the coverage of adsorbates,[9,10] while RDS is more sensitive to the surface anisotropy.[11] A theoretical study indicated that layer-by-layer oxidation on Si(001) should be detectable with RDS.[12] Recently, Yasuda *et al.* have shown that it is possible to detect layer-by-layer oxidation as an RD oscillation,[13] and they also evaluated the activation energies for the growth of two or more oxide layers on Si(001) from the RD oscillation period.[4]

It has been theoretically proposed that the initial interaction of an oxygen molecule on the buckled dimer of Si(001) predominantly provides adsorption of oxygen at the backbond sites through barrierless dissociation.[14] At a temperature below 473 K, the barrierless process was experimentally supported by an AES study combined with scanning reflection electron microscopy (SREM).[15] However, it appears to be still an open question whether a finite activation energy exists even at higher temperatures. We show here that the activation energy can be estimated from RD oscillation measurements during the growth of an oxide monolayer on Si(001) at high temperatures. An early UPS study found no significant changes of O(2*p*) uptake curves even above 623 K.[16] Their results suggested that there is no substantial activation energy, although this point was not explicitly discussed. On the contrary, our results presented here show that

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a finite activation energy exists for monolayer oxidation above 473 K.

II. EXPERIMENT

Experiments were performed with an ultrahigh vacuum chamber, whose base pressure was under 2.0×10^{-8} Pa, as measured with an ionization gauge. The sample used was a *p*-type Si(001) single crystal with resistivity of 12.0–14.0 Ω cm, cut to the size of $3\times20\times0.63$ mm^3. We used a Si(001) wafer with a miscut at 4° toward the [110] direction to acquire single-domain Si(001)-(2×1) structure. Heating was performed by applying direct current to the sample. The sample was well degassed for over 12 h at 873 K and cleaned by flashing at 1470 K for 10 s. It was then subjected to oxidation with molecular O_2 at a pressure of 1.0×10^{-5} Pa in the temperature range of 583–923 K. We used a cold-cathode gauge to monitor the oxidation pressure in order to avoid electron-stimulated effects arising from the hot filament of the ionization gauge. The substrate temperature was calibrated by means of direct attachment of a K-type (alumel-chromel) thermocouple to the sample in the same heating setup. The relation between the sample temperature (T_s) measured with the thermocouple and the heating current (I) followed the equation $I=A(T_s-T_0)^B$ over the temperature range of 583–823 K, where T_0 represents room temperature. An infrared pyrometer was also used to calibrate the sample temperature above 823 K. In RD measurements, a thermocouple was not directly attached to the sample to avoid Ni contamination. It should be noted that the signal-to-noise ratio is better at higher temperatures because the heating current is larger and more stable.

The optical setup for the RD measurements was designed following the configuration reported by Aspnes *et al.*[17] We used a strain-free window to minimize optical anisotropy irrelevant to the surface structure. A xenon lamp was used as the light source from the visible to near-ultraviolet region (2.0–5.0 eV). The incident light polarized in the [010] direction was introduced almost normal to the surface. Reflected light was monochromated with a monochromator located in front of the photomultiplier.

The RD amplitude, $\Delta r/r$, is defined as

$$\frac{\Delta r}{r}=\frac{2(r_a-r_b)}{r_a+r_b}, \tag{1}$$

where r_a and r_b are complex reflectances for polarization parallel and perpendicular to the direction of the dimer bonds on a clean Si(001)-(2×1) surface, respectively. The real part of the RD amplitude $[\Delta R/R=\mathrm{Re}(\Delta r/r)]$ was exclusively analyzed since the real part and the imaginary part are connected via the Kramers-Kronig relation.

III. RESULTS

In Fig. 1, we present typical RD spectra obtained for a clean surface and an oxidized surface. Here, the surface was oxidized at the oxygen pressure of 1.0×10^{-5} Pa and at the temperature of 583 K for 20 min. Spectral features of the clean surface are consistent with those reported in the literature.[18,19] A shoulderlike structure at 3.4 eV, corresponding to the E_1 and E_0' critical points, appears to be a fingerprint of a single-domain Si(001)-(2×1) surface with a 4° miscut angle.[18] This feature is ascribed to the surface modified bulk optical transition at the L and Γ points in the Brillouin zone, respectively. A pronounced peak structure around 3.1 eV for a clean surface at room temperature is characteristic of a surface with high step density.[18] In this paper, we present time courses at a fixed photon energy of 3.1 eV since a main component of the RD spectra is considered to appear at around 3.1 eV at high temperatures, being slightly redshifted from the E_1 critical point of 3.4 eV.[4]

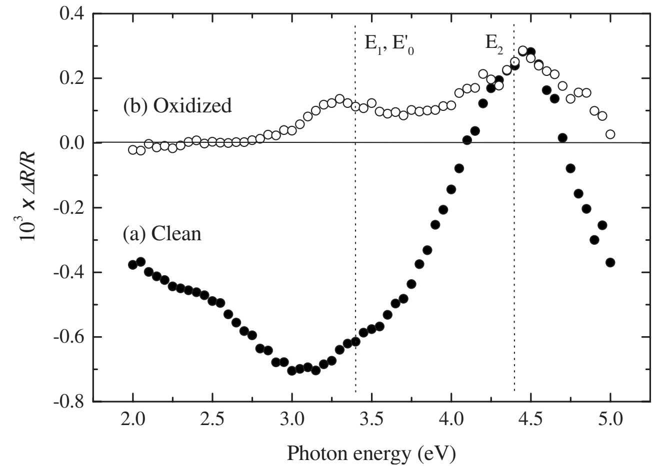

FIG. 1. (a) RD spectrum of a clean Si(001)-(2×1) surface with 4° miscut toward the [110] direction. (b) RD spectrum of the surface oxidized at the oxygen pressure of 1.0×10^{-5} Pa and at the temperature of 583 K for 20 min.

Typical time courses of RD intensity for the initial stage of oxide growth on Si(001)-(2×1), i.e., monolayer oxidation, are shown in Fig. 2. Two temperatures were selected to present the results for different growth modes. The spectral line shape of the oxidized surface in Fig. 1(b) is a typical feature of the RD spectra at the near saturation point in the present temperature range. At higher oxidation pressure and temperature, further oxidation in the layer-by-layer mode for the growth of two or more oxide layers was observed as the RD oscillation.[4] Here, we present the initial part of the RD oscillation, corresponding to the monolayer oxidation.

The RD intensity will be decreased by further oxidation to form a second oxide layer.[4] In RD measurements, we observed a slight increase of the RD intensity even a long time after it reached near saturation. It is possible to discriminate the rapid oxidation regime and the slow oxidation regime under the present oxidation conditions. In real-time AES, a linear increase of the total oxygen coverage in the slow oxidation regime was interpreted as oxidation occurring at an oxide interface after the completion of an oxide monolayer.[5] It is possible that growth of a second oxide layer might begin at this stage. Hence, we consider here that the end of the rapid growth corresponds to the completion of an oxide monolayer. After subtraction of the linear increase in the slow oxidation regime, the normalized time courses can be fitted with a single exponential function as follows:

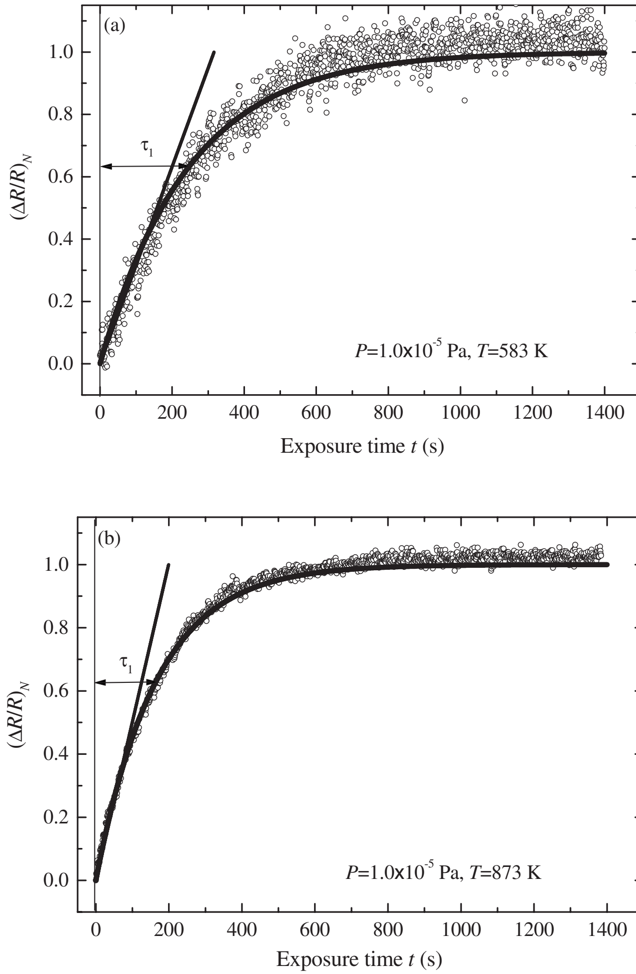

FIG. 2. Time courses of RD intensity at 3.1 eV measured *in situ* during oxidation at a pressure of 1.0×10^{-5} Pa. Lines represent linear fitting of the plots for $\Delta R/R \leqslant 0.3$. The growth modes are (a) Langmuir-type adsorption at 583 K and (b) two-dimensional island growth at 873 K.

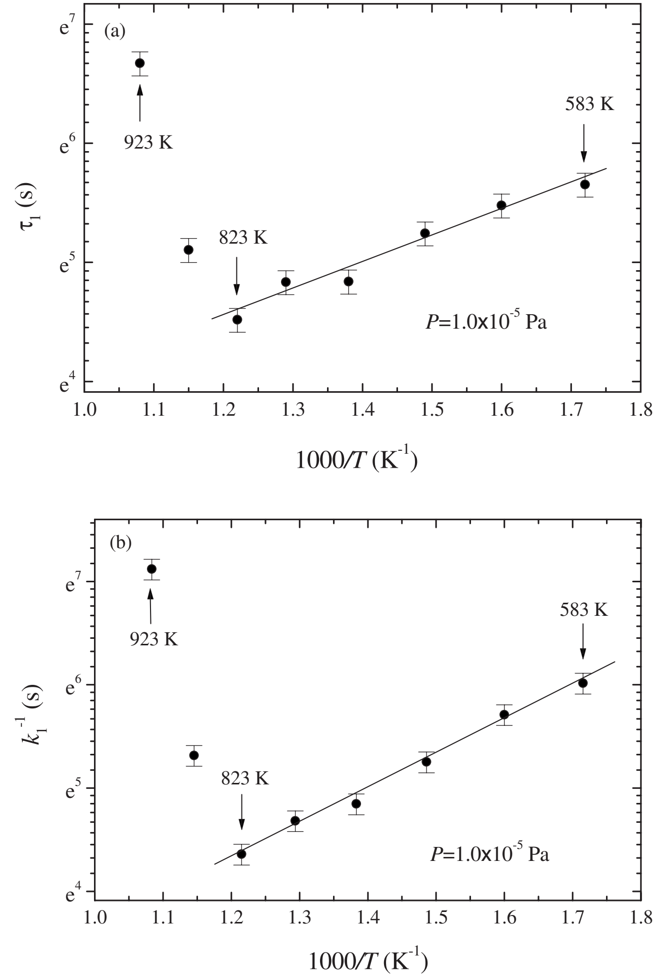

FIG. 3. (a) Arrhenius plots for τ_1 measured at a pressure of 1.0×10^{-5} Pa. The fitted line corresponds to the Langmuir-type adsorption regime below 823 K. (b) Arrhenius plots for k_1^{-1} obtained for $\Delta R/R \leqslant 0.3$ at a pressure of 1.0×10^{-5} Pa. The fitted line corresponds to the Langmuir-type adsorption regime below 823 K.

$$(\Delta R/R)_N = 1 - \exp\left(-\frac{t}{\tau_1}\right), \tag{2}$$

where N denotes the normalization of the intensity, t represents the oxidation time and τ_1 corresponds to the time constant as presented in Fig. 2. The uptake curves are normalized so that the intensity converges to 1 at the end of the rapid oxidation regime. A similar fitting was performed in real-time AES analysis.[5] In a recent RDS study, Yasuda *et al.* used the following expression for the growth of the nth oxide layer:

$$\frac{1}{\Delta t_n} = C \exp\left(-\frac{\epsilon_n}{k_B T_s}\right), \tag{3}$$

where Δt_n is the oxidation time during the growth of the nth oxide layer in layer-by-layer mode, C is postulated to be constant with temperature but can be dependent on the oxidation pressure, k_B is the Boltzmann constant, and ϵ_n is postulated to be the energy barrier for the rate-governing process for each layer.[4,20] To obtain the activation energy, we utilize τ_1 in place of Δt_1 in the present analysis. It should be noted that the RD oscillations do not follow a simple curve for the growth of two or more oxide layers.[4]

We also performed fitting with a linear function to obtain the initial slope k_1 for the data plots of $(\Delta R/R)_N \leqslant 0.3$. These k_1 values are used as the initial slope of $(\Delta R/R)_N$. It is possible that these k_1 values would give a different activation energy. The value of ϵ_1 obtained from fitting of an exponential function should be considered as estimating the *averaged* value of the activation energy in terms of the oxygen coverage. On the other hand, the value of ϵ_1^* should be interpreted as representing the activation energy at low coverage.

IV. DISCUSSION

The obtained oxidation periods (τ_1) at various temperatures are plotted in Arrhenius form in Fig. 3(a). A minimum value of τ_1 appears at 823 K. In the Langmuir-type adsorption regime (583–823 K), the value of τ_1 decreases with temperature, while in the two-dimensional island growth

TABLE I. Activation energies in layer-by-layer oxidation. Asterisk represents the activation energy at the initial stage.

Oxide layer i	Activation energy ϵ_i (eV)	Oxidation conditions P (Pa)	T (K)	Method
First	$\epsilon_1=0.16\pm0.03$	1.0×10^{-5}	583–823	This work
First	$\epsilon_1^*=0.26\pm0.03$	1.0×10^{-5}	583–823	This work
First	$\epsilon_1=0.0$	2.7×10^{-5}	623–827	UPS[a]
First	$\epsilon_1=0.0$	1.3×10^{-5}	RT–473	AES[b]
First	$\epsilon_1^*=0.0$			Calc[c]
Second	$\epsilon_2=1.2\pm0.3$	0.01–20	952–1200	RDS[d]
Second	$\epsilon_2^*=0.3$	2.7×10^{-4}	643–908	AES/SREM[b]
Third	$\epsilon_3=2.0\pm0.2$	0.06–20	952–1200	RDS[d]
Fourth	$\epsilon_4=2.2\pm0.2$	2.0–20	952–1200	RDS[d]

[a]Reference 16.
[b]Reference 15.
[c]Reference 14.
[d]Reference 4.

(823–923 K), τ_1 increases with temperature because a SiO desorption process is involved.[5] The minimum value of 823 K corresponds to the transition temperature between Langmuir-type adsorption and two-dimensional island growth. The obtained value of 823 K is slightly lower than the value extracted from AES data by Takakuwa *et al.*[5] It has been shown that the transition temperature between active oxidation and passive oxidation increases with oxidation pressure.[8] Our results may indicate that this trend is also the case for the transition temperature between Langmuir-type adsorption and two-dimensional island growth. In our previous study with SDR, we demonstrated that the SDR signal is proportional to the total oxygen coverage above 820 K.[9] The transition temperature obtained with SDR at the same oxidation pressure of 1.0×10^{-5} Pa is between 820 and 920 K and consistent with the present results.

The oxygen uptake curves with SDR exhibit different line shapes, i.e., a sigmoid curve of two-dimensional island growth and a single exponential curve of Langmuir-type adsorption.[9] On the other hand, such different line shapes were not observed with RD measurements across the transition temperature, as shown in Fig. 2. This clear discrepancy of the line shape from AES (Ref. 5) and SDR (Ref. 9) studies above 823 K indicates that $(\Delta R/R)_N$ from RD measurements is not proportional to the total oxygen coverage in the two-dimensional island growth regime. The line shape obtained with RD measurements below 823 K is consistent with the AES study of O(KLL) intensity,[5] as well as our previous SDR results.[9] At temperatures of 630–882 K, a recent UPS study clearly showed a single exponential increase of O($2p$) intensity.[21] In a recent study, change of RD intensity was postulated to be proportional to the coverage of adsorbates for various molecules at room temperature.[18] Here, we postulate that RD intensity is roughly proportional to the total oxygen coverage in the Langmuir-type adsorption regime below 823 K. The origin of the RD intensity in oxidation process will be addressed later.

We obtained an activation energy of $\epsilon_1=0.16\pm0.03$ eV from Arrhenius plots in Fig. 3(a). This value is compared with the reported activation energies, including those for the growth of two or more oxide layers, in Table I. Our results show the existence of a finite activation energy in monolayer oxidation at the temperature of 473–823 K at an oxygen pressure of 1.0×10^{-5} Pa. This result is at odds with the early UPS study, which showed that O($2p$)-derived valence band intensity curves exhibit no temperature dependence between 623 and 827 K at an oxygen pressure of 2.7×10^{-5} Pa.[16] According to an AES study combined with SREM, AES intensity curves showed no significant change below 473 K, supporting a barrierless process.[15] If a barrierless dissociation process is dominant, as proposed based on theoretical calculations,[14] no significant temperature dependence would be observed. However, such a picture is currently still a matter of debate. Recent scanning tunneling microscopy (STM) investigation on a stepped Si(001)-$c(4\times2)$ surface at 80 K suggested that mobile oxygen species with a long lifetime should exist on the terrace and migrate until they are adsorbed near the step edges.[22] This result may indicate that there would be a finite activation energy for oxidation to occur at the terrace. A recent theoretical study revealed that there may exist an activation energy for oxygen molecules to convert from a weakly bound physisorbed precursor state to a more stable chemisorbed state when incoming oxygen molecules do not take the geometry parallel to the dimer.[23] Based on the energy curves of singlet and triplet states, they derived a barrier height of 0.05 eV for the tilted oxygen molecule interacting with the up-dimer Si atom and that of 0.11 eV for the tilted oxygen molecule interacting with the middle point of the dimer. These values are comparable with the activation energy obtained in the present study and suggest that the barriers can indeed be accessed with RD measurements.

Kinetic effects involving migration of Si atoms at high temperatures should also be taken into account as the origin

of a finite activation energy. Our previous work with SDR showed a faster increase of the uptake curve for the component ascribed to the backbond adsorption of oxygen at room temperature.[9] This result remains consistent with the barrierless dissociation picture and preferential adsorption of oxygen at the backbond site.[14] The oxidation mechanism may be very different above and below 473 K, and the Si migration process appears to be activated above this temperature, as suggested by STM findings.[24] A recent first-principles theoretical study showed that the oxygen-adsorbed structure with one missing Si dimer may be energetically the most stable at low coverage.[25] Si dimer ejection may be a thermally activated process, though this was not addressed in the theoretical work. A more stable oxide structure can be formed through activation of mobile Si species. It is reasonable to suppose that the activation energy at high temperatures above 473 K would be slightly greater than the values of 0.05–0.11 eV theoretically obtained without including such kinetic effects. Considering our present results, it is still possible that barrierless dissociation process is dominant at temperatures below 473 K. Since a finite activation energy is currently suggested even at low temperatures on Si(001)-$c(4\times2)$, it may not be adequate to attribute its origin solely to kinetic effects. It is clear that construction of a theoretical framework including dynamic effects and temperature dependence is needed to assess activation energy in a monolayer regime.

To investigate the coverage dependence of activation energy, we analyzed the initial part of the time courses of RD intensity. Arrhenius plots using the data plots satisfying $(\Delta R/R)_N \leq 0.3$ are shown in Fig. 3(b). The transition temperature between Langmuir-type adsorption and two-dimensional island growth is similar to that in Fig. 3(a). The activation energy at low coverage below 823 K is estimated to be $\epsilon_1^* = 0.26 \pm 0.03$. The obtained value ϵ_1^* is larger than ϵ_1. The results indicate that a finite activation energy should exist even at low coverage. As summarized in Table I, the activation energy increases with the oxygen coverage for growth above the second oxide layer. On the other hand, the activation energy decreases with the oxygen coverage in the monolayer regime. It would be important to consider kinetic effects to assess the coverage dependence of the activation energy. When the dimer is attacked by an oxygen molecule and ejected, the next step of oxidation would be different from the oxidation on a flat Si(001)-(2×1) surface. In other words, interaction of oxygen molecules with a dimer vacancy might explain the reduction of the activation energy with increasing oxygen coverage in the monolayer regime. For two-dimensional island growth above 823 K, the desorption process of SiO should also play a role in the formation of the SiO_2/Si interface.[5] It has been pointed out that strain relaxation involving Si migration toward the SiO_2 film is a critical process to maintain a smooth SiO_2/Si interface above the monolayer regime.[26] Involvement of mobile Si or SiO species is a key to understand the oxidation process in various temperature ranges and at various coverages, not limited to a monolayer regime.

In a recent theoretical study of the RDS features of oxidized Si(001)-(2×1), changes of RD intensity at around the E_1 (E_0') and E_2 critical points were ascribed to stretching and/or compression deformation of the silicon lattice rather than to the electronic states of oxygen.[2] This interpretation means that it would be difficult to relate the obtained activation energy to specific adsorption sites. Other first-principles calculations for a monolayer oxide regime also showed that large RD intensity is produced around the interface between the oxide and the silicon substrate.[27] With the aid of theoretical insights, it should be possible to relate activation energy to the oxidation process including lattice relaxation. Thus, the values of ϵ_1 and ϵ_1^* might be considered as average values of the activation energy over various adsorption sites. Although the oxidation process at a specific adsorption site cannot be identified, the obtained activation energies with RDS may correspond to the rate-governing process as postulated for the growth of two or more oxide layers.[4]

Finally, RDS may be useful in combination with SDR to analyze site-dependent chemical processes in real time. The backbond site and the dimer interstitial site are considered to be the predominant adsorption sites at this stage.[28] The adsorption of oxygen bridging the neighboring dimer could also be a possibility.[29] Our previous study showed the feasibility of using SDR to discriminate simultaneous adsorption processes at different adsorption sites.[9] It is possible that SDR is more sensitive to the local adsorption structures, while RDS probes the relaxation of the whole lattice due to oxidation. Accordingly, we expect that the combination of RDS and SDR would be effective to clarify the activation energies toward different adsorption sites and the dynamic effects, involving both oxygen and silicon mobile species, in monolayer oxidation on Si(001)-(2×1).

V. SUMMARY

The initial oxidation process of the Si(001)-(2×1) surface was investigated by means of RDS. Analysis of the initial time courses of the RD intensity enabled us to estimate the transition temperature between two growth modes, namely, Langmuir-type adsorption and two-dimensional island growth. Furthermore, we found that a finite activation energy exists for monolayer oxidation at high temperatures. Our results also indicate that a finite activation energy should exist even at low coverage.

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