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Selective Chain Reaction of Acetone Leading to the Successive Growth of Mutually Perpendicular Molecular Lines on the Si(100)-(2×1)-H Surface

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Abstract: The successive growth of mutually perpendicular molecular lines from one dangling-bond (DB) site on the Si(100)-(2×1)-H surface has been realized through a substrate-mediated chain reaction at 300 K. Among various molecules, acetone molecules undergo the most facile chain reaction with a DB site, which proceeds selectively on the Si(100)-(2×1)-H surface, resulting in only single molecular lines in the parallel-row (parallel to the dimer row) direction. The smaller size and higher reactivity of acetone molecules enable us to successively grow a parallel-row acetone line from the end of a cross-row (perpendicular to the dimer row) allylmercaptan line simply by changing the feed of gas molecules into the reaction chamber. Since the length of a molecular line is controlled by the number of gas molecules impinged, it is possible to turn a chain reaction from the cross-row direction to the parallel-row direction at any desired point on the surface. The reaction path of the adsorbing molecules is discussed. The present study provides a new means of fabricating mutually perpendicular molecular lines through a chain reaction initiating at a preselected DB site on the Si(100)-(2×1) surface.

Introduction

The controlled assembly of organic molecules on silicon surfaces is of growing interest because of its potential application in molecular electronics, in which single molecules are expected to act as gating, switching, and other types of electronic devices.[1-4] One of the key challenges to the realization of future molecular devices is to integrate those functional individual molecules into a circuit. The substrate-mediated radical chain reaction of some selective molecules, initiated by a dangling-bond (DB) site on the H-terminated Si(100)-(2×1) surface, has emerged as one of the most promising approaches to fabricating a molecular line (adsorbed molecules assembled in one dimension) connecting two desired points on the surface.[1,5-9] The Si(100)-(2×1)-H surface consists of mono-hydride silicon dimer rows running parallel to each other, where the interdimer distance across rows (7.68 Å) is twice that (3.84 Å) along a row. Hence, to fabricate a molecular line connecting two predefined points not lying on the same symmetric axis on the Si(100)-(2×1)-H surface, it is necessary to direct a self-directed chain reaction, initiated at a preselected DB site, in both parallel-row (parallel to the dimer row) and cross-row (perpendicular to the dimer row) directions successively. The success in steering a chain reaction in two perpendicular directions will lead to the fabrication of desired line structures of molecular assemblies on the Si(100)-(2×1)-H surface.

To date, a number of studies have demonstrated molecular line formation with a variety of organic molecules on the Si(100)-(2×1)-H surface through the chain reaction mechanism.[1,5-10] Except for the allylmercaptan (ALM) molecule,[8] organic molecules such as styrene,[1,7] vinyl ferrocene,[5] and benzaldehyde[10] show the growth of molecular lines in the parallel-row direction. Recently, we have observed that the DB-initiated chain reaction of ALM molecules proceeds in the cross-row direction, forming an ALM line on the Si(100)-(2×1)-H surface.[8] The basic reaction steps of molecular line growth with various molecules are similar to those reported by Linford et al. for hydrocarbon adsorption on the Si(111) surface in a liquid environment.[11] Although the reaction proceeds through the

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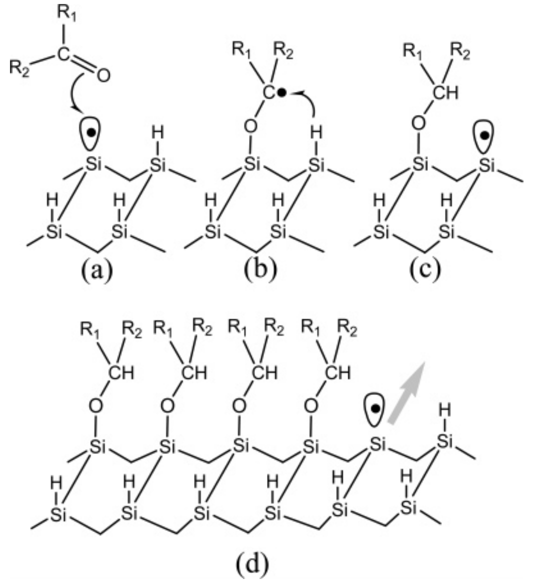

Figure 1. (a−c) Schematic of a generalized mechanism of the parallel-row chain reaction of a molecules having a >C═O moiety with a dangling bond (DB) on the Si(100)-(2×1)-H surface. (d) DB site at the end of a parallel-to-dimer-row molecular line. The arrow indicates the cross-row (perpendicular to the dimer-row) direction.

interaction of the C═C or C═O part of a molecule with a DB of the substrate, many molecules containing a C═C or C═O moiety show no molecular line growth.[1,6,9] Hence, in spite of the successful growth of molecular lines with aldehyde (acetaldehyde and benzaldehyde),[10] one cannot be certain that a ketone will show molecular line growth on the Si(100)-(2×1)-H surface in a similar fashion.

The generalized mechanism of a parallel-row chain reaction involving a DB site and a C═O moiety is shown schematically in Figure 1a−c.[10] The C-centered intermediate radical (Figure 1b) abstracts a H from the nearest dimer in the same row to generate a new DB site (Figure 1c), which can react with another molecule and repeat the reaction steps (a−c). Thus, a free DB always exists at the end of a freshly grown molecular line (end-DB), as shown in Figure 1d, unless it is terminated or quenched by some means, such as reaction with a H atom or any pre-existing defect.[1] Similarly to an isolated DB site on the surface, the end-DB of a parallel-row (or cross-row) molecular line should initiate a chain reaction in the cross-row (or parallel-row) direction when appropriate molecules are dosed onto the Si(100)-(2×1)-H surface. To date, there have been no studies, to our knowledge, that succeeded in growing perpendicularly connected molecular lines through a chain reaction initiated from one DB site on the surface.

In this study, we report the successive growth of two mutually perpendicular molecular lines through a chain reaction initiated from one DB site on the Si(100)-(2×1)-H surface at 300 K. We observed that acetone molecules, among the various molecules that have been studied to date, undergo the most facile chain reaction with a DB site on the surface very selectively and form only single acetone lines in the parallel-row direction. The selective and facile reactions of acetone molecules lead us to turn a chain reaction, initiated by an isolated DB site, from the cross-row direction to the parallel-row direction on the Si(100)-(2×1)-H surface. Thereby, a junction of a cross-row ALM line and a parallel-row acetone line was fabricated without creating any additional DB site on the surface. The contrast of an acetone line, observed in STM images, depends on the bias voltage applied during scanning, which helps us to understand the reaction path of adsorbing molecules. The present findings concerning a new kind of molecule (ketone) undergoing a chain reaction and a means of fabricating mutually perpendicular molecular lines from one DB site are expected to promote further studies that will lead to the fabrication of desired junctions of molecular lines on the silicon surface.

Experimental Section

The experiments were performed in an ultra-high-vacuum chamber with a base pressure lower than 6×10^{-11} mbar. A variable-temperature scanning tunneling microscope (Omicron VT-STM) was used. The B-doped silicon sample (0.01 Ω·cm) was cleaned by prolonged annealing at ~850 K (~8 h), followed by repeated flashing at 1400 K. The clean surface was H-terminated by exposure to atomic H, which was generated by a hot W filament (~2100 K), at a surface temperature of ~625 K. The freshly prepared H-terminated surface usually contains a dilute concentration of unpaired DBs resulting from incomplete H-termination;[12] otherwise, a DB at a predefined position is generated using the STM tip.[13] Acetone (dehydrated) and allylmercaptan (ALM) were purchased from Tokyo Chemicals Industries Co., Ltd. (TCI) and Wako Chemicals, respectively. Those colorless liquid chemicals were further purified by several freeze−pump−thaw cycles or by pumping out the vapor above the liquid phase. The molecules were dosed onto the silicon surface through an electronically controlled pulse-valve doser. The number of molecules dosed is expressed in langmuir units ($1\ \mathrm{L} = 1 \times 10^{-6}$ Torr·s), which is estimated from the background pressure reading by BA gauge during the exposure without any correction for the ionization coefficient.

Results

Figure 2 shows a series of STM images of the same area on the Si(100)-(2×1)-H surface at 300 K. The surface shown in Figure 2a, which contains a few DB sites indicated by arrows, was used as the initial surface for this series of measurements. The parallel stripes running diagonally in each terrace are monohydrated silicon dimer rows. In addition to some irregularly shaped defect sites and pre-adsorbed molecules, a pre-existing molecular line was also observed on the surface. Note that the surface was exposed to some gas molecules, including acetone, before present controlled experiment. When the surface shown in Figure 2a was exposed to ~0.02 L of acetone molecules, several molecular lines initiated at DB sites were observed, as shown in Figure 2b. By inspecting hundreds of molecular lines, we found that acetone molecules always form single molecular lines (as observed in Figure 2b). Unlike styrene and other molecules that form molecular lines in the parallel-row direction,[1,6,10,14] acetone molecules never form double lines where both Si dimer atoms in a row are occupied by adsorbed molecules. In rare events, only one or two underlying Si dimers were observed to be doubly occupied by acetone molecules in a molecular line.

It was observed that the molecular lines formed by adsorbed acetone molecules are normally stable during scanning in the

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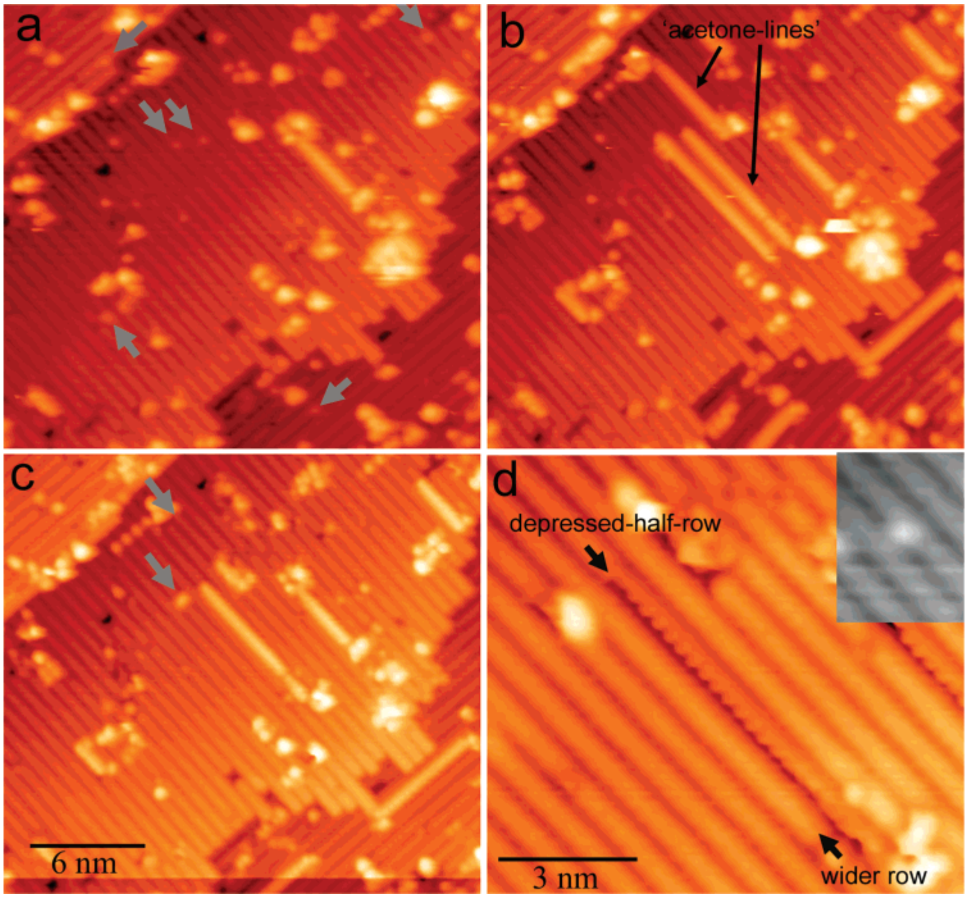

Figure 2. Selected area STM images of the Si(100)-(2×1)-H surface showing growth of molecular lines of adsorbed acetone molecules through the chain reaction mechanism at 300 K. (a) Initial Si(100)-(2×1)-H surface containing several DB sites, indicated by arrows. In addition to those DB sites, the surface also contains several defects and some pre-adsorbed molecules. (b) Surface shown in panel a exposed to ~0.02 L of acetone molecules at room temperature. (c) Surface shown in panel b after scanning at positive sample bias. Arrows indicate the dimer rows from which molecular lines disappeared. (d) Selected area of surface shown in panel c scanned at −1.6 V sample bias. The inset shows the location of a DB site in a dimer row. $V_{\mathrm{sample}} = -2.3$ V (a−c); $I = 0.2$ nA (a−d).

occupied state at sample biases below ~4.0 V. At higher scanning biases (> −5 V), the molecular lines were decomposed (data not shown). However, when a surface containing several freshly grown molecular lines was scanned in the unoccupied state, even at lower biases (for example, at +2 V), some of the molecular lines disappeared, leaving a clean H-terminated dimer row, as observed in Figure 2c. As mentioned earlier, a freshly grown molecular line should have an end-DB. This end-DB can initiate a chain reaction, similar to a forward reaction, in the reversed direction to desorb molecules in a line.[14] The main indication of desorption through the chain reaction mechanism is the re-appearance of a H-terminated dimer row in an STM image after the desorption of molecules from a molecular line, as observed in Figure 2c. It seems that the scan at a positive sample bias (scanning in the unoccupied state) facilitates the reverse chain reaction in the present case. The role of tunneling electrons in the desorption process has not yet been clarified.

The appearance of molecular lines of adsorbed acetone molecules in an STM image strongly depends on the bias voltage applied during scanning. As shown in Figure 2d, the molecular lines appeared as depressions at a lower sample bias of −1.6 V, whereas they appeared as bright protrusions at −2.3 V (Figure 2a−c). Note that the threshold voltage for changing the appearance of a molecular line depends on the atomic structure of the STM tip apex. Careful inspection of Figure 2d reveals that one half of the dimer row appeared as a depression (indicated by a depressed half-row), while the other half was similar to a clean H-terminated dimer row. In addition, the dimer row adjacent to the depression side appeared wider (indicated in Figure 2d) than a row in clean terrace. The depressed half-row in Figure 2d lies on the same side of the dimer row at which the initial DB site is located in Figure 2a (as shown in the inset).

In contrast to other molecules studied to date, ALM molecules undergo a chain reaction in the cross-row direction, resulting in an ALM line on the Si(100)-(2×1)-H surface.[8] Figure 3 shows a series of STM images of a selected area of the Si(100)-(2×1)-H surface, where Figure 3a was used as the initial surface for this series of measurements. In addition to the DB site indicated, a few other bright protrusions (defects) are observed in Figure 3a.[12] When the surface shown in Figure 3a was exposed to ~0.4 L of ALM molecules at 300 K, an ALM line grown from the DB site in the cross-row direction was observed, as shown in Figure 3b.[8] The black and blue arrows (Figure 3b) indicate the sites of the initiation and end of the growth of the ALM line, respectively. One can easily realize from the STM images shown in Figure 3a,b that the end of the ALM line lies in the clean area on the surface; i.e., the growth of the ALM line does not end at any pre-existing defect on the surface, which can quench a DB.[1] Hence, a free DB site should exist at the

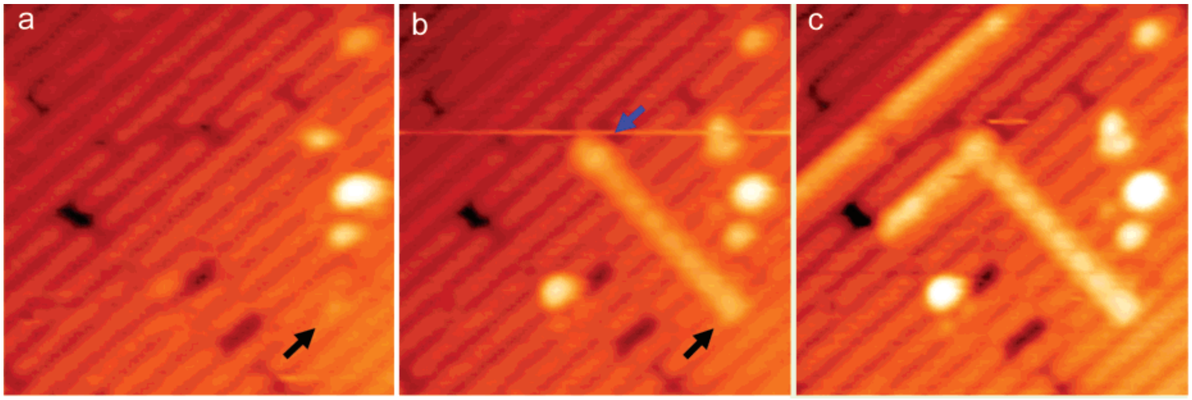

Figure 3. Successive growth of mutually perpendicular molecular lines by propagating a chain reaction from the cross-row direction to the parallel-row direction on the Si(100)-(2×1)-H surface at 300 K. (a) Initial Si(100)-(2×1)-H surface containing a dangling bond (DB) site, indicated by a black arrow. (b) Surface shown in panel a exposed to ∼0.4 L of allylmercaptan (ALM) molecules. The black and blue arrows indicate the initiation and end of ALM line growth, respectively. (c) Surface shown in panel b exposed to ∼0.2 L of acetone molecules. Scan area, 11 × 11 nm^2; $V_{\text{sample}} = -2.6$ V; $I = 0.2$ nA.

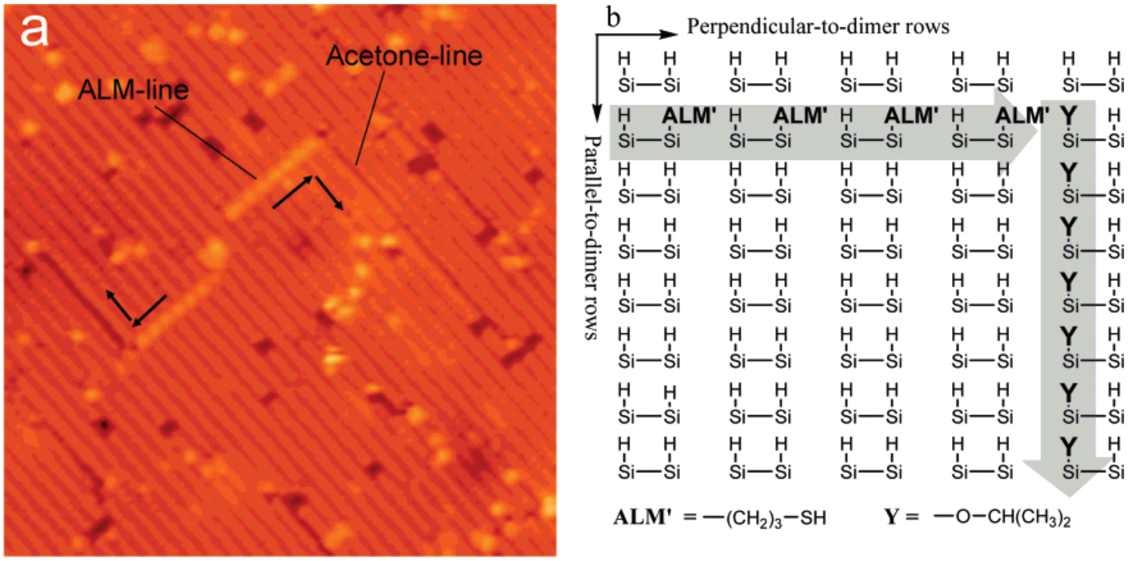

Figure 4. (a) Zoomed-in STM image of successively grown allylmercaptan and acetone lines on the Si(100)-(2×1)-H surface. $V_{\text{sample}} = -2.4$ V; $I = 0.2$ nA. The growth directions of the ALM and acetone lines are indicated by arrow. (b) Schematic alignment of acetone line formed through a chain reaction from end-DB of an ALM line on the Si(100)-(2×1)-H surface. Gray arrows indicate the growth directions of ALM and acetone lines.

end of the ALM line. When the surface shown in Figure 3b was exposed to ∼0.2 L of acetone molecules, a parallel-row molecular line appeared, initiated from the end of the ALM line (indicated by blue arrow), as shown in Figure 3c. The fabrication of such a molecular line from the end of a freshly grown ALM line, without creating any additional DB site, is well reproduced. In addition to the molecular line grown from the end of the ALM line, another molecular line in the parallel-row direction also appeared in Figure 3c. We also attempted to grow an ALM line from the end of a molecular line of adsorbed acetone but did not succeed.

Note that the end-DB of an ALM line (as indicated by blue arrows in Figure 3b) was generated through the abstraction of one of the two anisotropic H's from the next row by the intermediate radical of adsorbed ALM.[8] It can be realized from Figure 3b that it is hard to determine the abstraction site of H simply by looking into the ALM line growth, because the protrusion of end-DB overlaps with the nearest ALM molecule. However, by registering the molecular line grown from the end of an ALM line, we can determine the site from which H was abstracted by the adsorbed ALM intermediate.[8] Figure 4a shows a high-resolution STM image of the Si(100)-(2×1)-H surface, where parallel-row molecular lines were grown from the end of ALM lines by procedures similarly to those used in the case of Figure 3. The growth directions of the cross- and parallel-row molecular lines are indicated by arrows. The parallel-row molecular lines are observed as depressions in one half of a dimer row in this image, similar to those observed in Figure 2d. The analysis of several (∼10) junctions of such molecular lines revealed that the depressed half of the dimer row lies at the side facing the ALM line, as observed in Figure 4a. Incidentally, both of the parallel-row molecular lines in Figure 4a were grown to the right of the direction of ALM line growth; however, molecular lines grown to the left of the direction of ALM line growth were also observed, as seen in Figure 3c.

Discussion

Previous studies suggested that only a few selected molecules produce molecular lines on the Si(100)-(2×1)-H surface through a chain reaction initiated by a DB site.[6,8,14] It was also reported that only a fraction of DB sites on the Si(100)-(2×1)-H surface initiate a chain reaction to produce molecular lines.[1,8] The number of impinging molecules necessary to produce a typical molecular line varies with the kind of molecules dosed onto the surface; for example, the doses of benzaldehyde and acetaldehyde molecules needed for growing a molecular line were 5 and 120 L, respectively.[10] All these observations are thought to be mainly related to the stability of the intermediate radical (Figure 1b), which abstracts a H from a nearby dimer within a row to push the reaction in the forward direction.[6,14−16] The results shown in Figure 2 clearly demonstrate that acetone molecules undergo a chain reaction with a DB site and produce an acetone line in the parallel-row direction on the Si(100)-(2×1)-H surface, similar to styrene and aldehyde.[1,10] However, the dose of acetone molecules necessary (∼0.02 L) to produce a typical molecular line was much lower than that of other molecules (>1 L) using the same dosing system. This suggests that the chain reaction of acetone molecules with a DB site on the Si(100)-(2×1)-H surface is much facile compared with those of other molecules, such as styrene and acetaldehyde.[1,6,10]

The reaction steps involved for producing an acetone line are expected to be similar to those shown in Figure 1, where $R_1 = R_2 = CH_3$. The initial step is the interaction of $>C{=}O$ with a DB site of the substrate to form a C-centered intermediate radical (Figure 1b). The facile reaction of acetone molecules indicates that the C-centered intermediate radical of an adsorbed acetone molecule is more stable than other molecules that have been studied so far.[6,15,16] This enhanced stabilization of the C-centered radical of an adsorbed acetone molecule may be related to the electron-donating properties of methyl groups. However, some calculation is needed to explore the exact reason for and magnitude of the enhanced stability of an intermediate radical.

It has been reported that, like an isolated DB, the end-DB of a parallel-row molecular line can initiate a chain reaction with different molecules in the same direction, resulting in hetero-molecular lines on the surface.[17] However, the growth of a molecular line from an end-DB in the perpendicular direction, which is important to produce a junction of molecular lines connecting two points not lying on the same symmetric axis on the surface, has not been obtained so far. In a previous attempt to grow a 2,4-dimethylstyrene (and styrene) line from the end-DB of an ALM line, it was observed that a styrene molecule interacts with the end-DB but does not undergo a chain reaction to form a molecular line.[9] In fact, the adsorbed styrene was desorbed from the end-DB in a few minutes. Hence, to connect a pre-existing ALM line with a styrene line, an additional DB site had to be created using an STM tip.[9]

In the present study, the growth of an acetone line from the end of an ALM line (Figure 3) clearly demonstrated that there was a DB at the end of a cross-row ALM line, which initiated a chain reaction in the parallel-row direction when the surface was exposed to acetone molecules. The smaller size and more facile reaction of acetone molecules with a DB site, relative to those of styrene molecules, may be the reason for the successful growth of an acetone line from the end of an ALM line without creating any additional DB using an STM tip. These results suggest that a chain reaction initiated by a DB site can be easily directed from the cross-row direction to the parallel-row direction at any desired point on the surface by changing the feed of reacting gas molecules into the reaction chamber. Note that the length of both ALM and acetone lines can be controlled by controlling the number of gas molecules fed into the chamber.[1,8] The growth of an acetone line to either the left or the right of the direction of the ALM line growth indicates that, like that initiated by an isolated DB site, the chain reaction initiated by an end-DB of an ALM line can proceed in either direction along the dimer row.

The registries of the DB site and respective acetone line on an underlying dimer row (Figure 2) clearly indicate that the chain reaction of acetone molecules proceeds selectively on the same side of a dimer row where the DB is initially located. The depressed half of a dimer row at a lower sample bias (Figures 2d and 4a) can be ascribed to the adsorption of acetone molecules at one of the Si−Si dimer atoms through an O−Si bond. Because the electronegativity of the O atom is higher than that of the Si atom, an O-bonded Si atom is expected to show an electronic state different from that of the other Si atom of the dimer. Consequently, O- and H-bonded Si atoms of a dimer should appear differently in STM images, as observed in the bias-dependent measurement. However, details of the electronic states involved in the appearance of acetone lines with various biases are not clearly understood at the present stage. The wider appearance of a dimer row adjacent to the depressed half-row (Figures 2d and 4a) suggests that the $-CH(CH_3)_2$ part of an adsorbed acetone molecule lies between neighboring dimer rows; i.e., the $CH(CH_3)_2$ part is tilted away from the Si−Si dimer that reacted. The absence of any double lines of adsorbed acetone molecules indicates that the C-centered intermediate radical cannot approach the H atom on the same dimer (Figure 1b) to abstract it, possibly because of the steric hindrance suffered by the two methyl groups.

The location of the depressed half-dimer of an acetone line grown from the end-DB of an ALM line indicates (Figure 4a) that acetone molecules are bonded to the nearest Si atoms of the dimers in the row next to the end of an ALM line, as shown schematically in Figure 4b. Thus, the present study indicates that the end-DB of an ALM line is located at the nearest Si atom of the dimer row next to ALM lines; i.e., the intermediate radical of adsorbed ALM abstracts the nearest H from the next row (toward the growth direction), as proposed in our previous study.[8] Because our experimental observations are limited to only ∼10 junctions of ALM and acetone lines, we do not rule out the existence of DB on the other Si atom of the dimer.

In the present study, the growth of an acetone line from the end-DB of an ALM line is well reproduced. However, the attempts to grow an ALM line from the end-DB of an acetone line were not successful, which limits the formation of a more extended junction of molecular lines on the surface. The low probability of ALM molecules to undergo a cross-row chain reaction or the steric hindrance induced by the nearby adsorbed acetone molecule may be the reason for this. The observation of a facile reaction by acetone molecules (a new kind indeed) with a DB site is expected to promote further study to find additional molecules that can undergo a cross-row chain reaction, leading to the fabrication of more complex molecular line structures.

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Conclusions

The successive growth of mutually perpendicular molecular lines starting from one DB site on the Si(100)-(2×1)-H surface has been studied using STM at 300 K. We observed that acetone molecules undergo a very facile and selective chain reaction with a DB site on the Si(100)-(2×1)-H surface, forming molecular lines in the parallel-row direction. The facile reaction of acetone molecules with a DB site enables us to fabricate a junction of a cross-row ALM line and a parallel-row acetone line from one DB site at a predefined location. This result suggests that a chain reaction initiated by a DB site can be directed from the cross-row direction to the parallel-row direction at a desired point on the surface by changing the feed of reacting molecules into the chamber. Unlike other molecules, acetone molecules show exclusively single molecular lines, in which molecules are bonded to one of the Si−Si dimer atoms lying on the same side of a row; i.e., the chain reaction does not flip onto the other side in a dimer row. The registries of an acetone line on an underlying dimer row help us to understand the undergoing reaction paths of adsorbing molecules. The present findings are expected to promote further study to find more molecules that can undergo facile reactions in both directions, leading to the fabrication of more complex molecular line structures on the surface.

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