The reason why thin-film silicon grows layer by layer in plasma-enhanced chemical vapor deposition

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Thin-film Si grows layer by layer on Si(001)-(2×1):H in plasma-enhanced chemical vapor deposition. Here we investigate the reason why this occurs by using quantum chemical molecular dynamics and density functional theory calculations. We propose a dangling bond (DB) diffusion model as an alternative to the SiH3 diffusion model, which is in conflict with first-principles calculation results and does not match the experimental evidence. In our model, DBs diffuse rapidly along an upper layer consisting of Si-H3 sites, and then migrate from the upper layer to a lower layer consisting of Si-H sites. The subsequently incident SiH3 radical is then adsorbed onto the DB in the lower layer, producing two-dimensional growth. We find that DB diffusion appears analogous to H diffusion and can explain the reason why the layer-by-layer growth occurs.

Thin-film Si has attracted attention as a promising material for solar cells1. Plasma-enhanced chemical vapor deposition (PECVD) using SiH4 is a key technique for fast large-area growth at low temperatures2–5. Tsai et al. showed that SiH3 radicals are the predominant deposition precursors and effective for forming “device-quality” films, that is, films with atomically smooth surfaces2,3. Obtaining smooth surfaces is particularly important for practical applications because rough surface morphologies induce surface and interface defects and reduce carrier lifetimes. Elucidation of the mechanisms by which layer-by-layer growth of thin-film Si occurs in PECVD is therefore strongly desired for the development of high-performance solar cells.

Experimental studies of surface morphologies have suggested reasons behind the surface smoothing of thin-film Si3,6–11. It is well known that surface diffusion of deposition precursors is essential for explaining the surface smoothing mechanism in simple homoepitaxial growth using molecular beam epitaxy (MBE)12. Kukushkin et al. performed remarkable and important works on layer-by-layer growth mechanisms of thin-film in epitaxial growth13–16. They proposed the theory of layer-by-layer growth by diffusional coalescence of faceted islands both on a crystal surface and on an island containing screw dislocations. Then, they theoretically demonstrated that the tablet- and needle-like growth of thin-film take place when the main mode of mass-transport is two- and three-dimensional diffusion on the substrate, respectively. Furthermore, there is almost no doubt that surface diffusion produces smooth surfaces in CVD because experimental results have revealed a strong dependence of surface roughness on substrate temperatures and growth rates3,17. Matsuda et al. found a correlation between surface loss probabilities of SiH3 radicals and surface H coverage and suggested a three-center diffusion model11,17. In that model, SiH3 radicals are physisorbed onto Si-H sites and diffuse along the hydrogenated surface. Although the three-center diffusion model has been accepted to some extent, scaling behavior analyses of surface morphologies using atomic force microscopy (AFM) and in situ ellipsometry have not found physisorption sites for SiH3 radicals18. Additionally, diffusion models remain an indirect explanation of the smoothing mechanism because experimental observations of surface reactions at the atomic scale are difficult. Computational simulations are thus effective for gaining a direct understanding of the surface reactions in PECVD. Maroudas et al. suggested a “valley-filling” mechanism based on molecular dynamics (MD) simulations19. They referred to microscopic higher- and lower-deposition areas as surface hills and valleys, respectively, and suggested that SiH3 radicals diffuse from surface hills to valleys and preferentially passivate dangling bonds (DBs) located in surface valleys. They argued that SiH3 radicals are weakly adsorbed on the hydrogenated surface. However, Cereda et al. used density functional theory (DFT) calculations to show that SiH3 radicals abstract H atoms by overcoming the negligible small activation energy20. This means that SiH3 radicals cannot exist on a hydrogenated surface in the weakly physisorbed state. Long-distance diffusion of weakly physisorbed SiH3 radicals is therefore unlikely. We
also found from tight-binding quantum chemical MD simulations that the initial growth of thin-film Si follows an “abstraction-adsorption” mechanism\(^{21,22}\), where a DB is generated on the surface via H abstraction by a SiH\(_3\) radical and the next SiH\(_3\) radical is adsorbed onto the generated DB. Thus, although experimental results have found a relationship between surface diffusion and layer-by-layer growth\(^{23,24}\), the diffusion mechanism of SiH\(_3\) radicals has not yet been confirmed, and the previously proposed diffusion mechanisms thus remain controversial. In particular, the questions of what species diffuses and how the surface diffusion affects the formation of atomically smooth surfaces are still open to debate. Against this background, we perform self-consistent-charge density-functional tight-binding (SCC-DFTB)\(^{25,26}\) MD and DFT calculations and report a surface diffusion mechanism that can explain how smooth surfaces are formed by Si PECVD. An understanding of the layer-by-layer growth mechanism is expected to enable atomic-scale design of PECVD processes for producing thin-film Si with atomically smooth surfaces leading to low defect densities and high photostability.

SCC-DFTB MD simulations of a Si(001)-(2\(\times\)1):H surface were performed by continuous impingement of 25 SiH\(_3\) radicals at intervals of 2.0 ps. We calculated 15 trajectories for durations of up to 50 ps. Figure 1 shows snapshots of a CVD simulation of a typical trajectory. On the Si(001)-(2\(\times\)1):H surface, an incident SiH\(_3\) radical abstracts a H atom and generates a SiH\(_4\) molecule (Fig. 1(a) and (b)). A DB is also generated on the surface. The subsequently incident SiH\(_3\) radical is then adsorbed onto the DB (Fig. 1(c) and (d)). This is in agreement with our previously proposed “abstraction-adsorption” mechanism\(^{22}\). At this point, we define the terms lower layer and upper layer for referring to the layers of Si atoms terminated by H atoms. The lower layer is defined as the top layer of Si atoms of the original Si(001)-(2\(\times\)1):H surface, and the upper layer is defined as the layer above the lower layer and consists of Sih\(_2\) sites generated by the abstraction-adsorption process. After the first 24.0 ps of simulation time, four SiH\(_3\) radicals have been adsorbed onto the surface via the abstraction-adsorption mechanism. Next, an incident SiH\(_3\) radical abstracts a H atom from an adsorbed SiH\(_3\) species (Fig. 1(e) and (f)). The reaction generates a Si-H\(_2\) site with a DB in the upper layer (Fig. 1(g)). The Si-H\(_2\) site abstracts a H atom from a neighboring Si-H site in the lower layer. This effectively means that the DB migrates from the upper layer to the lower layer (Fig. 1(h)). The Si-H\(_3\) site thus re-forms via the DB migration, with a DB newly generated in the lower layer. Over the course of 15 MD trajectories, DB migration from the upper layer to the lower layer is observed six times, while migration from the lower layer to the upper layer is observed only once. Another incident SiH\(_3\) radical is then adsorbed onto the DB located in the lower layer (Fig. 1(i) and (j)). The next incident SiH\(_3\) radical abstracts a H atom from an adsorbed SiH\(_3\) species, again generating a Si-H\(_2\) site with a DB in the upper layer (Fig. 1(k)). The Si-H\(_2\) site abstracts a H atom from a neighboring Si-H\(_2\) site. The DB migrates from the Si-H\(_2\) site to a neighboring Si-H\(_3\) site in the same layer (Fig. 1(l)). We find that DB migration can be classified into three patterns: (1) from a Si-H\(_2\) site in the upper layer to a Si-H site in the lower layer; (2) from a Si-H\(_2\) site in the lower layer to a Si-H\(_3\) site in the upper layer; and (3) from a Si-H\(_2\) site in the upper layer to a Si-H\(_2\) site in the same layer. These patterns of DB migration occur in 23%, 4%, and 73% of the 15 MD trajectories, respectively. However, we do not observe physisorption or surface diffusion of SiH\(_3\) radicals, though these phenomena were previously suggested by experimental groups\(^{27}\). Our simulation shows that physisorbed SiH\(_3\) radicals do not migrate whereas DBs frequently do so on the hydrogenated surface.

To validate the migration of DBs from a Si-H\(_2\) site in the upper layer to a neighboring Si-H site in the lower layer on Si(001)-(2\(\times\)1), which corresponds to pattern (1) described above, we calculated the activation energies of three possible migration paths from a step site using DFT. In Fig. 2(a), P1, P2, and P3 represent the migration of a DB between two adjacent dimer rows, between two adjacent dimers in the same dimer row, and within the dimer, respectively. Figure 2(b) shows that the activation energies of P1, P2, and P3 are 8.29, 11.66, and 19.27 kcal/mol, respectively. However, the reverse reactions of P1, P2, and P3 require higher activation energies of 15.65, 19.02, and 26.63 kcal/mol, respectively, which correspond to
pattern (2). This means that DBs stay in the lower layer for long periods of time. Moreover, our DFT calculations show that it is difficult for DBs to diffuse along the lower layer, which consists of Si-H sites, due to the high activation energy of 43.85 kcal/mol, and such diffusion is not observed in our SCC-DFTB MD simulations. The DBs thus remain at step sites in the lower layer. This indicates that the next SiH$_3$ radical could be adsorbed onto a step site in the lower layer.

Next, we calculated the activation energies for the migration of DBs from a Si-H$_2$ in the upper layer to a Si-H$_3$ site in the same layer via three possible migration paths corresponding to pattern (3). In Fig. 3(a), P4, P5, and P6 represent the migration of a DB between two adjacent dimer rows, between two adjacent dimers in the same dimer row, and within the dimer, respectively. Our DFT calculations yield activation energies of 1.37, 4.47, and 9.08 kcal/mol for P4, P5, and P6, respectively. These values are much smaller than those for migration from the upper layer to the lower layer (see Fig. 2). This means that DBs migrate more rapidly within the upper layer than from the upper layer to the lower layer. This is in good agreement with our SCC-DFTB MD calculations.

Our calculations find that, in addition to the adsorption of SiH$_3$ radicals via an abstraction-adsorption mechanism$^{22}$, the DBs on the hydrogenated surface exhibit three important behaviors: (a) DBs migrate from a Si-H$_2$ in the upper layer to a Si-H$_3$ site in the same layer via three possible migration paths corresponding to pattern (3). In Fig. 3(a), P4, P5, and P6 represent the migration of a DB between two adjacent dimer rows, between two adjacent dimers in the same dimer row, and within the dimer, respectively. Our DFT calculations yield activation energies of 1.37, 4.47, and 9.08 kcal/mol for P4, P5, and P6, respectively. These values are much smaller than those for migration from the upper layer to the lower layer (see Fig. 2). This means that DBs migrate more rapidly within the upper layer than from the upper layer to the lower layer. This is in good agreement with our SCC-DFTB MD calculations.

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Experimental results have shown that the surface diffusion of DBs during the interval between impingements of SiH$_3$ radicals on the upper layer strongly influences surface roughness$^{16,17}$. To reduce the surface roughness, we thus need to investigate how long the DBs can...
According to transition-state theory, the time derivative of the existence probability \( \theta_{ij} \) (0 ≤ \( \theta_{ij} \) ≤ 1) of a DB at site \( P_{ij} \) is given by the sum of the inflows from adjacent sites and outflows to adjacent sites:

\[
\frac{d\theta_{ij}}{dt} = \sum_{m} k_{ij}^{m} \theta_{im} - \sum_{m} k_{ij}^{m} \theta_{jm}, \quad i = 1, N; \quad 2 \leq j \leq N - 1,
\]

where \( m \) is an index that related to an adjacent site, and \( N \) is the length of each side in the calculation model. For \( 2 \leq i, j \leq N - 1 \), the site belongs to the upper layer; otherwise, the site belongs to the lower layer. The top equation corresponds to the derivatives for sites in the upper layer, and the second and third equations correspond to the derivatives for sites in the lower layer. These are integrated numerically using a fourth-order Runge-Kutta method for a maximum of 0.1 s. We consider only the case of an odd number \( N \). Since the generation of DBs can be expected to be random in the upper layer, we assume the existence probabilities of DBs at all sites in the upper layer are equal at \( t = 0.0 \) s; that is, \( \theta_{2,2} = \theta_{3,3} = \cdots = \theta_{ij} = \cdots = \theta_{N-1,N-1} = \frac{1}{(N-2) \times (N-2)} \) (2 ≤ \( i, j \leq N - 1 \)). At \( t = 0.0 \) s, the existence probabilities at all sites in the lower layer are equal to 0. The total probability for all sites including both the lower and upper layers is equal to 1 at \( t = 0.0 \) s, and this is conserved during the numerical calculations. The calculations are terminated when the total probability for all sites in the lower layer reaches 0.9. We define the time until the calculation terminates as the time needed for DB diffusion from the upper layer to the lower layer. Figure 5(b) shows the time needed for the total probability of the lower layer to reach 0.9 as a function of substrate temperature and \( N \). The dependency on \( N \) is important because SiH₃ island size increases with time during actual CVD growth. First, we focus on the behavior of the existence probability at low temperatures (less than 700 K). The time needed for the total probability of the lower layer to reach 0.9 increases as the size of the SiH₃ island.

**Figure 4** Schematic diagram of layer-by-layer growth of thin-film Si by PECVD. (a) Generation of a DB via H abstraction of a SiH₃ radical. (b) Adsorption of a SiH₃ radical on a DB. (c) DB diffusion within the upper layer consisting of SiH₃ sites. (d) DB diffusion from the upper layer to the lower layer. (e) Generation of a DB in the lower layer. (f) Two-dimensional growth by the adsorption of SiH₃ radicals onto the lower layer.
increases and as the substrate temperature decreases. To check the validity of our calculations, we compare the diffusion length obtained from our numerical calculation results with that obtained from experimental results. The diffusion length strongly depends on the time interval between incident SiH₃ radicals reaching the upper layer because DBs have to diffuse from the upper layer to the lower layer before the next SiH₃ radical impinges on the upper layer in order for lateral growth to occur. We estimate the diffusion length using both the necessary time calculated above and the time interval obtained from experimental data on SiH₃ radical densities in plasma. Experimentally, the cavity ring down technique for detection of SiH₃ radicals found high SiH₃ radical densities of 2.0 × 10¹⁹ to 1.2 × 10²⁰ m⁻³. From these results, we estimate that SiH₃ radicals reach the surface every 1.8 × 10⁻⁸ s per area of 10.0 × 10.0 nm² at a SiH₃ radical density of 1.0 × 10¹⁹ m⁻³ at 400 K. The area of 10.0 × 10.0 nm² is almost equal to the SiH₃ island size of N = 29 considering that there is one Si-H₃ site per area of 0.384 × 0.384 nm² on Si(001) surface. Figure 5(b) shows that the necessary time is about 1.8 × 10⁻⁸ s at 400 K when N = 29, which corresponds approximately to a time interval of 4.4 × 10⁻⁷ s per area of 20.0 × 20.0 nm². The average diffusion length thus increases to 5.0–10 nm at 500 K. Collins et al. showed that the diffusion length is between 6 and 10 nm at about 500 K by real-time spectroscopic ellipsometry, which is in good agreement with our results. Next, we focus on the behavior of existence probabilities at high temperatures (greater than 800 K). In Fig. 5(b), the lines at 800 and 900 K stop at N = 43 and 27, respectively, which indicates that the existence probabilities did not reach 0.9 within 0.1 s. The behavior of the existence probabilities at greater than 800 K is significantly different from that at less than 700 K. In Fig. 5(b), the time needed for the total probability of the lower layer to reach 0.9 at greater than 800 K is separated into four regions by inflection points and discontinuity with respect to N. In Region I, DBs diffuse rapidly from the upper layer to the lower layer because the reaction of P3 is likely to occur at high temperatures. This again reduces the slopes. In Region IV, although the
reactions are the same as in Region III, the existence probabilities in the upper layer increase because it is difficult for the DBs to reach the step edges in the large SiH$_3$ island, and the DBs thus remain on the upper layer. Therefore, at 900 K, the existence probability in the lower layer is unable to reach 0.9 when $N = 29$. As a consequence, an interesting phenomenon occurs where the necessary time increases discontinuously between $N = 27$ and 29. Our results indicate that the increase in substrate temperatures causes the return of DBs to the upper layer, which decreases the existence probability of DBs in the lower layer, because DBs stay on the upper layer for a long period of time when the SiH$_3$ island size is large. As Figure 5(b) shows, the DBs start to return to the upper layer at 800 K, a phenomenon that becomes more pronounced as the substrate temperature increases. This results in the adsorption of SiH$_3$ radicals onto the upper layer and an increase in surface roughness. Kondo et al. used AFM to show that surface roughness increases with increasing substrate temperatures, which is consistent with our calculation results. Taken together, our results show how DBs diffuse a long distance and what substrate temperatures are suitable for obtaining atomically smooth surfaces.

In PECVD processing of thin-film Si, it was previously believed that the SiH$_3$ radical deposition precursors were the diffusing species. This was based on the fact that the precursors of Si adatoms diffuse on the surface during MBE deposition. However, this is inconsistent with DFT calculation results and has no supporting experimental evidence. In this article, DBs are found to be the diffusing species in PECVD. Flewitt et al. studied the evolution of the surface topography using in situ scanning tunneling microscopy and proposed that preferential creation of DBs at step sites produces smooth surfaces, which is consistent with our results. However, they could not explain the long-distance diffusion of DBs, that is, how DBs on the terraces arrive at step edges. In contrast, the DB diffusion mechanism proposed in this article is able to explain the overall process of layer-by-layer growth.

In summary, we have used SCC-DFTB MD and DFT calculations to elucidate the reason why thin-film Si grows layer by layer in PECVD. DBs diffuse rapidly along the upper layer, which is covered with Si-H$_3$ sites. When the DBs reach step edges, they move down an atomic step. Owing to the longer residence time of the DBs at step sites in the lower layer, the next SiH$_3$ radical is adsorbed onto a step site in the lower layer, leading to lateral growth. We propose that this DB diffusion mechanism explains the layer-by-layer growth in Si PECVD.

Methods
We employ the SCC-DFTB MD method to simulate the CVD growth dynamics of thin-film Si at a finite temperature. The detailed methodology of SCC-DFTB is described elsewhere. The Si(001)-(2 × 1) surface has a slab geometry, and consists of 6 Si layers with 16 atoms per layer. The top and bottom layers are terminated with 32 H atoms. The slab model has a vacuum region of 22.7 Å. The temperature in SCC-DFTB MD simulations is set at 500 K. A time step of 0.1 fs is used. Fractional orbital occupations following the Fermi-Dirac distribution are employed. Undamped sources of deposited atoms. Acta metall. mater. 41, 1237–1241 (1993).

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**Author contributions**

T.K. and M.K. planned and supervised the study, and developed the original concept. T.K. developed the simulator and performed the simulations and data analysis. T.K. and Y.H. designed the theoretical model. T.K., H.I., K.K., Y.H., N.O. and M.K. discussed the results. T.K. wrote the manuscript, and Y.H. and M.K. revised the manuscript.

**Additional information**

**Competing financial interests:** The authors declare no competing financial interests.

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