

Supporting Information for

Deposition of hydrogenated silicon clusters for efficient epitaxial growth

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M1. Movie demonstrating a typical deposition of the amorphous $\text{Si}_{15}\text{H}_{10}$ cluster on a H-terminated Si(100)-(2×1) substrate with an impact energy of 5.30 eV/atom and a substrate temperature of 373 K under normal incidence.

M2. Movie demonstrating a typical deposition of the crystalline $\text{Si}_{29}\text{H}_{24}$ cluster on a H-terminated Si(100)-(2×1) substrate with an impact energy of 3.67 eV/atom, a substrate temperature of 473 K, and an incidence angle of 30°.

Parameter modification of the Ohira-Tersoff potential

The Ohira-Tersoff potential, V , is given as a function of the atomic coordinates by

$$V = \frac{1}{2} \sum_i \sum_{j \neq i} [a_{ij} V_r(r_{ij}) + b_{ij} V_a(r_{ij})] f_c(r_{ij}), \quad (1)$$

where

$$V_r(r_{ij}) = A_{ij} \exp(-\lambda_{ij} r_{ij}), \quad V_a(r_{ij}) = -B_{ij} \exp(-\mu_{ij} r_{ij}), \quad (2)$$

$$a_{ij} = \epsilon_{ij} (1 + \beta_i^{n_i} \tau_{ij}^{n_i})^{-1/2n_i}, \quad b_{ij} = \chi_{ij} (1 + \beta_i^{n_i} \xi_{ij}^{n_i})^{-m_i/2n_i}, \quad (3)$$

$$\tau_{ij} = \sum_{k \neq i, j} f_c(r_{ik}) \delta_{ik} g(\theta_{ijk}), \quad (4)$$

$$\xi_{ij} = \sum_{k \neq i, j} f_c(r_{ik}) \omega_{ik} g(\theta_{ijk}) \exp[\sigma_{ik}(r_{ij} - r_{ik})], \quad (5)$$

$$g(\theta_{ijk}) = 1 + \frac{c_i^2}{d_i^2} - \frac{c_i^2}{[d_i^2 + (h_i - \cos \theta_{ijk})^2]}, \quad (6)$$

$$f_c(r_{ij}) = \begin{cases} 1, & r_{ij} \leq R_{ij} \\ \frac{1}{2} + \frac{1}{2} \cos\left(\frac{\pi(r_{ij} - R_{ij})}{S_{ij} - R_{ij}}\right), & R_{ij} < r_{ij} < S_{ij} \\ 0, & r_{ij} \geq S_{ij} \end{cases} \quad (7)$$

In Eqs. 1-7, i , j and k denote the atoms in the system, r_{ij} is the distance between atoms i and j , and θ_{ijk} is the angle between the vectors \mathbf{r}_{ij} and \mathbf{r}_{ik} . The functions $V_r(r_{ij})$ and $V_a(r_{ij})$ in Eq. 2 represent a repulsive and an attractive pair potential, respectively. The smooth cutoff function $f_c(r_{ij})$ presented in Eq. 7 is employed to limit the range of the potential, and therefore, to reduce the computational cost. The functions a_{ij} and b_{ij} in Eq. 3 represent a measure of the bond order and strength, which depend on the local environment. The parameters (shown in Table 1) for Si-Si interactions were defined by Tersoff [1, 2], whereas the corresponding parameters for H-H, Si-H, and the corresponding three-body interactions were derived from *ab initio* calculations (configuration interaction methods), from a different empirical interatomic potential for Si-H interactions and from experimental measurements by Ohira and co-workers [3, 4, 5]. Table 2 shows the re-optimized S parameter for H-H interaction used in this work.

Table 1: Parameters of the Ohira-Tersoff potential.

| $i - j$ | Si-Si | H-H | Si-H, H-Si |
|-----------------------------|----------------------|--------|----------------------|
| $A(eV)$ | 1.8308×10^3 | 80.07 | 4.8733×10^2 |
| $B(eV)$ | 4.7118×10^2 | 31.38 | 1.8470×10^2 |
| $\lambda (\text{\AA}^{-1})$ | 2.4799 | 4.2075 | 2.9117 |
| $\mu (\text{\AA}^{-1})$ | 1.7322 | 1.7956 | 1.9898 |
| $R (\text{\AA})$ | 2.70 | 1.10 | 1.85 |
| $S (\text{\AA})$ | 3.00 | 1.7 | 2.05 |
| χ | 1.0 | 1.0 | 1.0485 |
| ϵ | 1.0 | 1.0 | 1.1027 |

| $i - j - k$ | Si-Si-Si | H-H-Si | Si-H-Si | H-Si-Si |
|-----------------------|----------------------|--------|----------------------|----------------------|
| $\sigma (\text{\AA})$ | 0.0 | 3.0 | 0.0 | 0.0 |
| c | 1.0039×10^5 | 0.0 | 1.0039×10^5 | 4.7762×10^5 |
| d | 16.217 | 1.0 | 16.217 | 6.3214×10^2 |
| ω | 1.0 | 4.0 | 0.0 | 0.6999 |
| δ | 0.0 | 0.0 | 0.0 | 0.0 |
| β | 1.1×10^{-6} | 1.0 | 1.1×10^{-6} | 1.0 |
| n | 0.78734 | 1.0 | 0.78734 | 1.0 |
| h | -0.59825 | -1.0 | -0.59825 | -1.0 |
| m | 1.0 | 1.6094 | 1.0 | 1.6094 |
| $i - j - k$ | Si-Si-H | H-H-H | Si-H-H | H-Si-H |
| $\sigma (\text{\AA})$ | 0.0 | 3.0 | 0.0 | 3.0 |
| c | 1.0039×10^5 | 0.0 | 1.0039×10^5 | 0.0 |
| d | 16.217 | 1.0 | 16.217 | 1.0 |
| ω | 1.5 | 4.0 | 0.4 | 4.0 |
| δ | 0.0 | 0.0 | 0.8512 | 0.0 |
| β | 1.1×10^{-6} | 1.0 | 1.1×10^{-6} | 1.0 |
| n | 0.78734 | 1.0 | 0.78734 | 1.0 |
| h | -0.59825 | -1.0 | -0.59825 | -1.0 |
| m | 1.0 | 1.6094 | 1.0 | 1.6094 |

Table 2: Reoptimized parameter of the Ohira-Tersoff potential.

| | |
|--------------------|------|
| $i - j$ | H-H |
| S (\AA) | 3.37 |

References

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