# Ordering Distance of Surface Nanofacets on Vicinal 4*H*-SiC(0001)

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After high-temperature $H_2$ etching, vicinal SiC(0001) surfaces showed periodically ordered nanofacet structures consisting of pairs of (0001) and $(11\bar{2}n)$. Here, we found that the characteristic ordering distance of ~10 nm is independent of the vicinal angle (4°–8° off). However, fluctuation in the ordering distance is dependent on the vicinal angle. The 5.7° off surface showed superior periodicity. The classical elastic theory of a surface predicted the characteristic (constant) ordering distance but not the fluctuation in ordering periodicity. By introducing ‘‘quantized step bunching’’ due to periodic surface energy, which is unique to polymorphic SiC, the fluctuation is described.

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Vicinal solid surfaces of single crystals often show regularly spaced step or terrace [1] and facet structures and thus have been considered as a template of nanostructures in heterosystems [2,3]. For future device applications, using the surface nanostructures physics behind self-ordering or self-organizational phenomenon is of great importance. In particular, equilibrium faceting, including step bunching on vicinal surfaces, is an important phenomenon that contributes to periodic surface morphology in a mesoscopic scale. This can be applicable to low-dimensional confinement systems such as quantum wires and dots in semiconductors. Surface faceting is observed in solid surfaces such as metals and semiconductors and is discussed based on kinetics and energetics [4–6]. Kinetic step bunching is typically observed during epitaxial growth of semiconductor thin films because of asymmetric diffusion of adatoms across a step, which produces a hill-and-valley surface morphology. Similar morphology can also be achieved via energetic processes. Surface phase separation [1] followed by ordering is responsible for this morphology. The classical linear elasticity theory by Marchenko [7] indicated a periodical faceting feature, where an ordering distance is energetically determined by minimizing the elastic relaxation energy at facet edges.

Silicon carbide is a unique compound semiconductor that possesses polymorphism. 3*C*, 4*H*, and 6*H* among more than ~250 polytypes are commercially available as a substrate and will be used for high power and high frequency electronic devices in the next generation [8]; in addition, they are strong candidates as a substrate for III-nitride epitaxy [9]. Chemically and structurally abrupt and defect-free interfaces are crucial for achieving high performance in these devices, making the surface of the SiC substrate of great importance. There have been many reports concerning surface cleaning and smoothing [10–15], possibly as a consequence of the rather rough and scratched features exhibited by commercially available SiC substrates [11]. High-temperature gas etching using $H_2$ [12–14], $H_2/HCl$ [11,15], and $H_2/C_2H_4$ [12,15] has been shown to be effective in attaining atomically smooth surfaces. Surface morphological studies have been conducted using these etched substrates, but are mostly viewed from the viewpoint of removal of surface defects for use in device applications. Nakamura *et al.* [11] reported step bunching on unintentionally vicinal SiC(0001) surfaces by etching kinetics and surface energies. We studied vicinal SiC(0001) surfaces (commercially available 3.5° off 6*H* and 8° off-axis 4*H*-SiC) after high-temperature $H_2$ etching and found self-organized nanofacet formation due to surface phase separation and self-ordering of nanofacets [16]. Energy considerations were provided, but the ordering mechanisms were not clear.

In this Letter, we shed light on the ordering mechanisms of nanofacets on vicinal 4*H*-SiC by investigating specially made SiC substrates having a series of vicinal angles. Nanofacet structures, ordering distances, and fluctuation in ordering at several vicinal angles are examined, and we provide a possible reason for the ordering using surface energetics.

4*H*-SiC(0001) substrates with vicinal angles of 4.2°, 4.9°, 5.7°, 6.4°, 7.2°, and 7.8° [all vicinal toward $[11\bar{2}0]$, Si face, and chemical-mechanical polish (CMP) treatment] were supplied by SiXON Ltd., Japan. They were initially cleaned with acetone, methanol, and 50% HF solutions. Samples were then etched by $H_2$ gas at 1360 °C for 30 min. Samples were examined *ex situ* by atomic force microscope (AFM) and high-resolution transmission electron microscope (HRTEM). Figures 1(a)–1(f) shows a typical image of the nanofacet surface at each vicinal angle. All images indicate ordered nanofacet features, as have been observed previously [16] and were confirmed by HRTEM in this study. Figures 2(a) and 2(b) show the cross-sectional TEM image of nanofacets and the corresponding schematic model. The nanofacet structure consists of a (0001) basal plane and a $(11\bar{2}n)$ ($n = 25$–30) nanofacet, inclined from (0001). The ordering distance observed in the AFM image is the length of a pair of (0001) and $(11\bar{2}n)$ nanofacet labeled as $L$ in Fig. 2(b). Note here that $h$, the height of

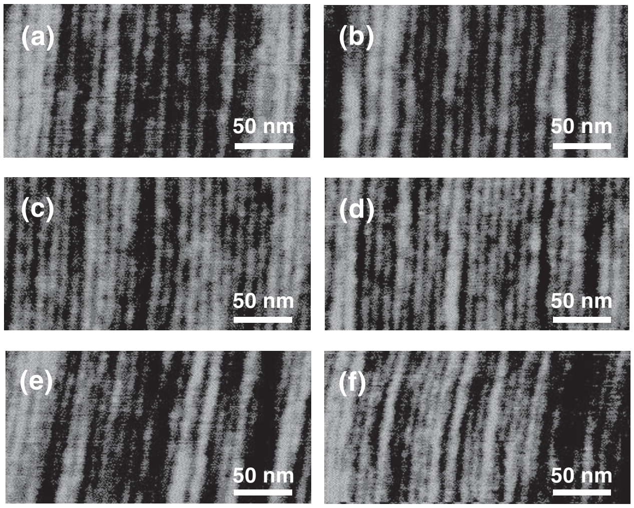

FIG. 1. Typical AFM images of (a) 4.2°, (b) 4.9°, (c) 5.7°, (d) 6.4°, (e) 7.2°, and (f) 7.8° off vicinal 4*H*-SiC (0001) surfaces after $H_2$ gas etching at 1360 °C. All surfaces indicate stripe patterns which correspond to the ordered nanofacet structure consisting of (0001) and $(11\bar{2}n)$ facets. Note that the degree of fluctuation of the ordering is minimized on the 5.7° off surface (c).

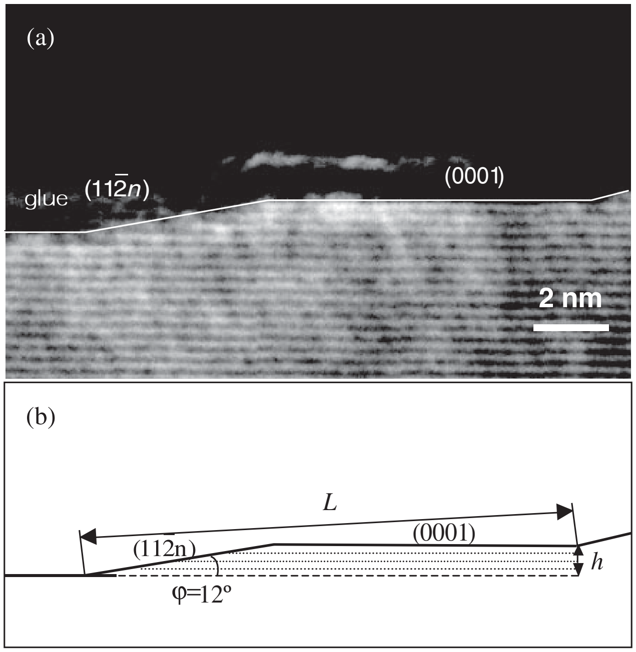

FIG. 2. (a) Cross-sectional TEM image of 5.7° off sample. Eye-guide lines are drawn to indicate the surface. (b) A model of the nanofacet structure on vicinal 4*H*-SiC. A nanofacet consists of a pair of (0001) and $(11\bar{2}n)$. Note the height (*h*) of a $(11\bar{2}n)$ facet is 1 nm, which is equivalent to a one-unit cell of 4*H*-SiC after "quantized step bunching."

$(11\bar{2}n)$ nanofacet, is predominantly 1 nm, which corresponds to four bimonolayer step heights and importantly is equivalent to a one-unit cell in the *c* axis of 4*H*-SiC. We sometimes observed more bunched $(11\bar{2}n)$ nanofacets whose height unexceptionally was *n* times a unit cell height (*n* is an integer) at relatively low etching temperatures [16], possibly due to metastable conditions. We call this "quantized step bunching" which occurs because of polymorphism in SiC. Statistical analysis of the ordering distance *L* using AFM images over the length of 10 $\mu$m (~1000 pairs of nanofacets) was performed. Figures 3(a)–3(f) indicate the distribution of the ordering distance at each vicinal angle surface. The results of peak separation are also indicated. The peak separation was performed using Gaussian fitting. We notice two important features from these results: First, the ordering distance peaks at ~10 nm, which is the so-called "characteristic ordering distance $L_0$," and is independent of vicinal angles possibly due to elastic effects on the surface [7], as seen in Fig. 4. Second, the degree of fluctuation of the ordering distance is at a minimum on the 5.7° off (vicinal) surface. The fact that there is a unique vicinal angle (5.7°) of 4*H*-SiC substrate reveals that highly ordered surface structures could in turn be a characteristic of polytypes of SiC. No such behavior has been observed in any other material.

We now consider the origins of the periodic surface nanostructures by hydrogen etching on vicinal SiC (0001) surface. We have reported that vicinal SiC(0001) surfaces are transformed into (0001) terraces and $(11\bar{2}n)$ nanofacets, which are organized to form hill-and-valley structures by pairing. The linear elastic theory of surface structures initially proposed by Marchenko and Parshin [7] predicts that such periodic nature of the surface is due to surface elastic energy minimization. The surface free energy per unit area *E* includes three different terms:

$$E = E_{\text{surf}} + E_{\text{edges}} + \Delta E_{\text{elastic}}, \qquad (1)$$

where $E_{\text{surf}}$ is the free energy of the (0001) terrace and $(11\bar{2}n)$ facet, $E_{\text{edges}}$ is the short-range energy of the edges, and $\Delta E_{\text{elastic}}$ is the elastic energy due to discontinuity of the surface stress at the crystal edges. These three terms are expressed as follows [17]:

$$E_{\text{surf}} = \frac{E_{(0001)}\sin(\varphi - \theta) + E_{(11\bar{2}n)}\sin(\theta)}{\sin(\varphi)}, \qquad (2)$$

$$E_{\text{edges}} = \frac{\varepsilon}{L}, \qquad (3)$$

$$\Delta E_{\text{elastic}} = -\frac{(1-\sigma^2)F^2}{\pi Y L}\ln\left[\frac{L}{\pi a}\sin\left(\pi\frac{\varphi-\theta}{\varphi}\right)\right]. \qquad (4)$$

Here, $E_{(0001)}$ and $E_{(11\bar{2}n)}$ are the surface energies of (0001) and $(11\bar{2}0)$, respectively, $\varepsilon$ is the short-range energy of the convex and concave edge, $\sigma$ is the Poisson's rate, *F* is the surface stress, *Y* is the Young's modulus, and *a* is the lattice parameter. When the surface free energy is minimized, by

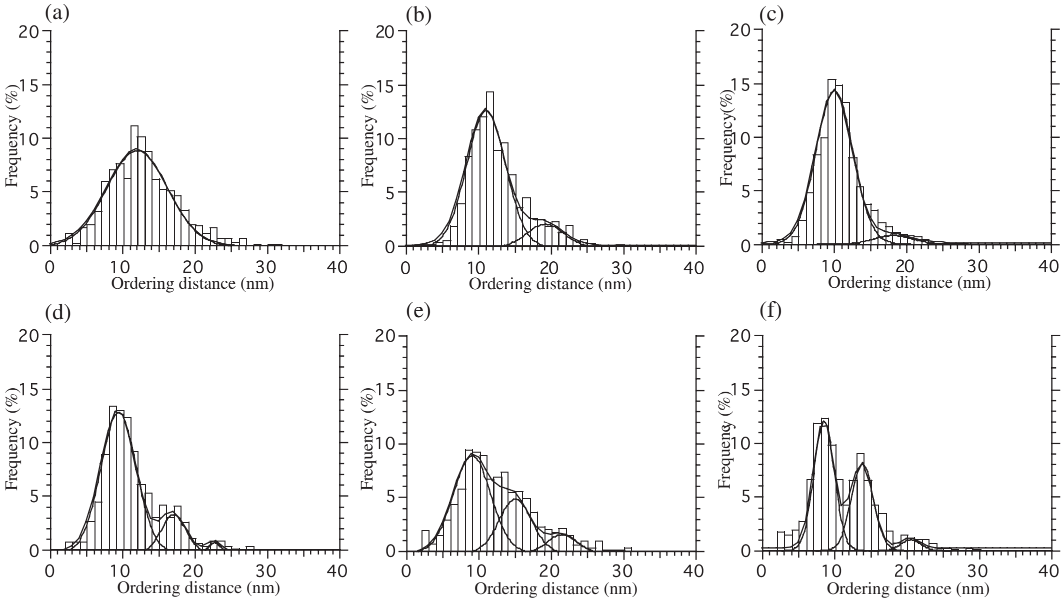

FIG. 3. Distribution of the ordering distance ($L$) at each vicinal angle surface of (a) 4.2°, (b) 4.9°, (c) 5.7°, (d) 6.4°, (e) 7.2°, and (f) 7.8° off samples based on statistical analysis of the AFM images. The results of peak separation are also indicated. Bold lines indicate fitting curves using each separated peak. Note that the main peak is ~10 nm independent of a vicinal angle, indicative of a characteristic ordering distance, and the degree of fluctuation of the ordering distance is at a minimum on the 5.7° off surface (c).

differentiating Eq. (1) with respect to $L$ and set $dE/dL = 0$, we obtain

$$L = \pi a \exp\left[\frac{\pi Y \varepsilon}{(1-\sigma^2)F^2}\right]\left[\sin\left(\pi\frac{\varphi-\theta}{\varphi}\right)\right]^{-1}. \quad (5)$$

This equation indicates that the ordering distance $L$ depends on the angle $\varphi$ between $(11\bar{2}n)$ and (0001) and the substrate vicinal angle $\theta$. In our results, the ordering (characteristic) distance $L$ is independent of the vicinal angle $\theta$; therefore, $\varphi = 2\theta$ is the condition to satisfy Eq. (5). Men *et al.* [18] reported the ordering distance after annealing in vacuum to be 63.5 nm, which differs from the distance of ~10 nm experimentally measured on vicinal SiC(0001) in our experiments. This could be attributed to an elastic parameter (surface stress) $F$ or an unknown change in materials, in Eq. (5). Thus far, it has been difficult to experimentally obtain the exact angle $\varphi$ at each substrate with different vicinal angles by analytical techniques. However, we found that the angle $\varphi$ is dependent on substrate vicinal angles $\varphi$ at smaller $\theta$ by *ex situ* reflection high energy electron diffraction. A Si (111) surface, which has crystallographically the same surface structure as SiC(0001), exhibits similar ordering behavior, the characteristic distance being independent of substrate vicinal angles. The elastic theory applied here could thus describe the presence of a characteristic ordering distance on vicinal SiC(0001) analog to Si(111). However, here, more attention and emphasis should be on the uniqueness of SiC polytypes that affect fluctuations in the ordering periodicity. As seen in Fig. 4, and according to theoretical considerations, the ordering distance is independent of substrate vicinal angles. However, the experimental results shown in the inset of Fig. 4 indicate that there is a slight deviation of the ordering distances from the characteristic distance of 10 nm. This behavior can be understood by introducing a periodic surface energy change in 4$H$-SiC(0001), which consists of $ABCB(A)$ stacking along the $c$ axis in one unit cell. According to surface energy

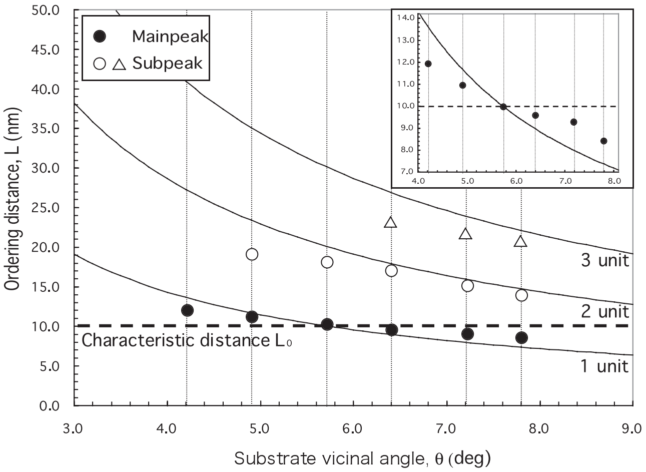

FIG. 4. Plots of the ordering distances after the peak separation. Calculated ordering distances based on quantized step bunching with $N$ ( = 1, 2, 3) unit. Note that the cross point of the characteristic ordering distance of 10 nm and the quantized step bunching curve at $N = 1$ gives a unique vicinal angle, resulting in superior surface ordering.

calculations using the Ising model [19,20], the surface energy of a stacking bilayer of $B$ is larger compared with other layers of $A$ and $C$ [21]. This partially supports our AFM results that indicate "quantized step bunching." However, the bunching height should be $cN/2$ ($c$ is a unit-cell height and $N$ is an integer) if this periodic energy change is applied. We speculate that other criteria may be present to stabilize nanofacets with $N$ unit-cell bunching.

Based on the $N$ unit-cell bunching criterion, the ordering distance at every vicinal angle ($\theta$) is geometrically given by $N$ and $\theta$ as

$$L = \frac{1.0}{\sin\theta} N. \tag{6}$$

Figure 4 shows the $L$-$\theta$ relation at different $N$ ($N = 1$, 2, 3), indicating a certain tendency of the deviation to center at $\theta = 5.7°$. From these results, we can infer that the ordering fluctuation is predominantly determined by two factors: quantized step bunching owing to the polytype nature of SiC and the characteristic ordering distance due to elastic effects under the vicinal angle conservation restriction. The cross point of the characteristic ordering distance of 10 nm and the quantized step bunching curve at $N = 1$ can thus give a unique (best for ordering) vicinal angle, which is calculated to be 5.74°. This could be the reason why the 5.7° off surface showed the superior periodicity.

In summary, we investigated vicinal SiC (0001) surfaces having a series of vicinal angles (4°–8° off) after high-temperature $H_2$ etching. Ordering of nanofacets, a pair of (0001) and $(11\bar{2}n)$, was observed on all surfaces. An ordering distance of ~10 nm was characteristically observed independent of vicinal angles, due to elastic energy minimization. Moreover, the degree of ordering fluctuation differed in each vicinal angle the dependence of the vicinal angle on periodicity. The elastic theory could predict a constant characteristic ordering distance but could not explain the fluctuation behavior. This is unique in SiC, which possesses polytypes; for example, 4$H$-SiC has a stacking sequence of $ABCB(A)$ along its $c$ axis, which results in quantized step bunching due to periodic surface energy. By combining the quantized step bunching originating from polytypic nature and the characteristic ordering distance the fluctuation in ordering and superior periodicity found in the 5.7° off surface could be well understood. The superiority at the 5.7° off surface implies significance in fabricating SiC-MOSFET or related devices since the surface or interface quality attributes to device performance.

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[1] H. C. Jeong and E. D. Williams, Surf. Sci. Rep. **34**, 171 (1999).
[2] A. Shchukin and D. Bimberg, Rev. Mod. Phys. **71**, 1125 (1999).
[3] H. Brune, M. Giovannini, K. Bromann, and K. Kern, Nature (London) **394**, 451 (1998).
[4] S. Rousset, F. Pourmir, J. M. Berroir, J. Klein, J. Lecoeur, P. Hecquet, and S. B., Surf. Sci. **422**, 33 (1999).
[5] M. Giesen and T. L. Einstein, Surf. Sci. **449**, 191 (2000).
[6] R. J. Phaneuf, N. C. Bartelt, E. D. Williams, W. Święch, and E. Bauer, Phys. Rev. Lett. **67**, 2986 (1991).
[7] V. I. Marchenko and A. Y. Parshin, Sov. Phys. JETP **52**, 129 (1980).
[8] *Silicon Carbide and Related Materials 2001*, edited by S. Yoshida, S. Nishino, H. Harima, and T. Kimoto (Trans Tech Publications, Zurich, 2002), Pt. 2.
[9] T. W. Weeks, Jr., M. D. Bremser, K. S. Ailey, E. Carlson, W. G. Perry, and R. F. Davis, Appl. Phys. Lett. **67**, 401 (1995).
[10] R. Yakimova, A. Hylén, M. Tuominen, M. Syvajarvi, and E. Janzen, Diam. Relat. Mater. **6**, 1456 (1997).
[11] S. Nakamura, T. Kimoto, H. Matsunami, S. Tanaka, N. Teraguchi, and A. Suzuki, Appl. Phys. Lett. **76**, 3412 (2000).
[12] A. Kawasuso, K. Kojima, M. Yoshikawa, H. Itoh, and K. Narumi, Appl. Phys. Lett. **76**, 1119 (2000).
[13] F. Dulot, L. Mansour, A. Leycuras, W. Wulfhekel, and D. Sander, Appl. Surf. Sci. **187**, 319 (2002).
[14] F. Owman, C. Hallin, P. Martensson, and E. Janzen, J. Cryst. Growth **167**, 391 (1996).
[15] Z. Y. Xie, C. H. Wei, L. Y. Li, Q. M. Yu, and J. H. Edgar, J. Cryst. Growth **217**, 115 (2000).
[16] H. Nakagawa, S. Tanaka, and I. Suemune, Phys. Rev. Lett. **91**, 226107 (2003).
[17] V. I. Marchenko, Sov. Phys. JETP **54**, 605 (1981).
[18] F. K. Men, F. Liu, P. J. Wang, C. H. Chen, D. L. Cheng, J. L. Lin, and F. J. Himpsel, Phys. Rev. Lett. **88**, 096105 (2002).
[19] C. Cheng, R. Needs, and V. Heine, J. Phys. C **21**, 1049 (1988).
[20] F. R. Chien, S. R. Nutt, W. S. Yoo, T. Kimoto, and H. Matsunami, J. Mater. Res. **9**, 940 (1994).

[21] According to Ref. [20] the surface energies of each stacking layer are follows: $E_A = E_C = 2.34$ meV, $E_B = 6.56$ meV.