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Pattern Formation via a Two-Step Faceting Transition on Vicinal Si(111) Surfaces

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Abstract

We demonstrate a self-organized pattern formation on vicinal Si(111) surfaces that are miscut toward the [$\bar{2}$11] direction. All the patterns, consisting of a periodic array of alternating (7×7) reconstructed terraces and step-bunched facets, have the same periodicity and facet structure, independent of the miscut angle; while the width of the facets increases linearly with miscut angle. We attribute such unique pattern formation to a surface faceting transition that involves two transition steps: the first step forms a stress-domain structure defining the universal periodicity; the second step forms the low-energy facets controlling the facet width.

Introduction

Surface patterning is an important processing step in many device fabrication processes. The continued drive to make devices smaller and smaller has brought up the challenge to pattern surfaces in the nanometer scale where conventional lithographic techniques are no long applicable. Two different routes have been taken toward nanopatterning: one by developing new patterning techniques with nanometer resolution, such as scanning probes, the other by taking advantage of self-organization of surface patterns occurring naturally. The later approach has shown great promises because it offers an economic and parallel process for device fabrication.

Surface stress often plays an important role in driving surface structural and morphological ordering, in particular, through the surface stress-induced spontaneous formation of ordered patterns of stress domains [1]. An effective way to create the stress-domain patterns is by step motion and/or by surface faceting transition on vicinal surfaces, as demonstrated on a variety of different materials surfaces, such as the Si(001) [2], Si(111) [3-5], GaAs(001) [6], Au(111) [7], and Pt (001) [8] surfaces.

Ideally, one would like to create a desirable surface pattern with controllable length scales. The characteristic length scales of the stress-domain patterns are generally determined by the competition between elastic relaxation energy and domain boundary energy. Therefore, in principle, it should be possible to control the length scales of such patterns by manipulating these energy terms. However, such kind of control is difficult to achieve in real practice. For example, the domain size of a stress domain can be changed by applying external strain [2], but the domain will restore its original size upon relieving the external force. On a vicinal surface, the terrace size can be changed by tuning miscut angle, but different step structures [9,10] and facets [5] usually form at different miscut angles.

Here we demonstrate the possibility of creating the same surface pattern of controllable length scales. We show that when the Si(111) surfaces are miscut toward the [$\bar{2}$11] direction,

thermal annealing leads to formation of one-dimensional surface patterns consisting of a periodic array of alternating flat terraces and high-step-density facets, with very good long-range order. Most important, all the surface patterns exhibit the same atomic structure for both the terraces and the facets and the same periodicity, independent of miscut angle; while the width of the facets increases linearly with increasing miscut angle. We propose that such unique self-organized pattern formation is achieved by a surface faceting transition that involves two transition steps. The first step, occurring at a higher temperature, is originated from a stress-induced instability [1,2]. It involves mass transport over the whole surface, forming tress-domain structures with a universal optimal periodicity and facet width (domain population) that minimize the surface strain energy. The second step, occurring at a lower temperature, is dominated by energy difference in surface reconstruction and facet structure. It involves mass transport in a shorter range within each period defined in the first step, forming the lowest-energy facet.

Experimental setup

Using scanning tunneling microscopy (STM), we have investigated the faceting behavior of Si(111) surfaces miscut toward the [$\bar{2}$11] direction over a wide range of miscut angles from 0.3° to 6°. The surfaces are cleaned by a standard procedure: degassing at 700°C for several hours in a vacuum chamber (base pressure 2×10^{-10} torr) followed by heating a few times to 1250°C for ~ 10 seconds each. The cleaned surfaces are then annealed at 950°C for a couple of minutes followed by slow cooling to room temperature at a rate of 1°C/sec. For each vicinality a large number of STM images are acquired from different Si(111) samples and from different cleaning-annealing cycles.

Figure 1 shows STM images of four typical ordered patterns formed on surfaces with a miscut angle of 1°, 2°, 4° and 6°, respectively, consisting of a periodic array of alternating (7×7) terraces and step-bunched facets. To quantitatively characterize the length scales of the patterns, the periodicity (terrace width + facet width) and the facet width are measured as a function of miscut angle. The value for a given miscut angle is derived from the peak position of the

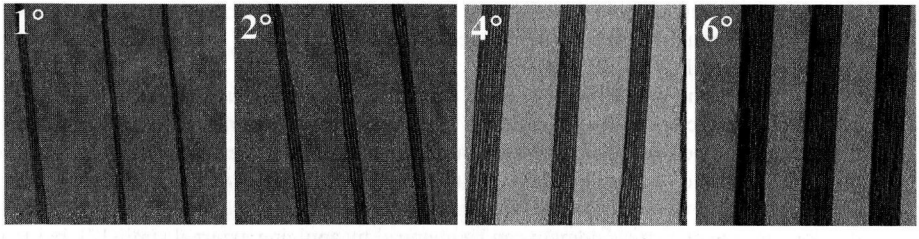

Fig. 1 Surface morphology of four well annealed vicinal Si(111) samples. The surfaces display an ordered pattern consisting of alternating (7×7) reconstructed terraces and high step-density facets. All four images have the same size of 2400 Å × 2400 Å. The downstairs direction runs from left to right. The derivative mode is used to emphasize the step structure in the facet.

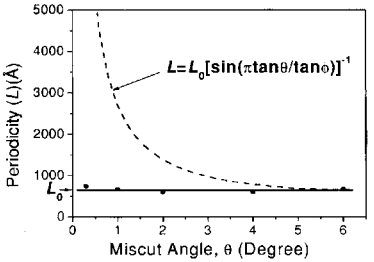

Fig. 2 Dependence of faceting periodicity on miscut angle. All the ordered patterns adopt a universal periodicity of ~635 Å, independent of miscut angle. Values (solid dots) for a given miscut are obtained from the peak positions of Gaussian fits to the measured data. The dashed line showed the dependence of periodicity on miscut angle if a one-step faceting transition had occurred (Please see text).

Gaussian distribution fit to the experiment data. The measured periodicities are 673, 593, 612, and 662 Å at miscut angles of to 1°, 2°, 4°, and 6°, respectively, with an average value of 635 Å.

All the patterns have the same periodicity independent of the miscut angle (Fig. 2); while the facet width increases linearly with miscut angle. Such a length-scale relationship implies that the facet in all the patterns must have the same facet angle, i.e., facet structure. This is indeed confirmed by high-resolution STM images of the facet. From those images we conclude that all the facets have an average step-step separation of 28 Å and a step height of 6.3 Å, amounting to a facet angle of 12.7°. The 6.3 Å equals to a double bi-layer step height in Si(111), so a transition from single bi-layer step to double bi-layer step must have occurred in addition to the bunching of steps during faceting. The same facet structure seems to be a special feature associated with azimuthal orientation of the $[\bar{2}11]$ direction. If the Si(111) surface is miscut toward other directions, different facets usually form at different miscut angles [5].

Discussions

We believe that these unique self-organized length-scale features are originated from a faceting transition in which surface stress plays an important role in addition to surface (facet) energy [3]. We propose that the periodicity and facet width is determined respectively by minimization of elastic energy and facet energy at two different steps of a faceting transition. Figure 3 shows schematically such a two-step faceting transition. At a temperature T_1 above transition, the surface consists of a staircase of (1×1) terraces separated by single bi-layer steps (Fig. 3a). As temperature is lowered below the transition temperature T_2, the surface starts nucleation of individual domains of the reconstructed (7×7) terraces and then self-organizes into

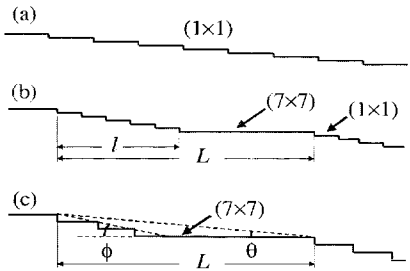

Fig.3 Schematics of a two-step faceting transition process. (a) Surface morphology above the thermodynamic phase transition temperature, consisting of equally spaced single bi-layer steps. (b) Morphology of a stress-domain structure formed below the first faceting transition temperature T_2, consisting of alternating of (7×7) reconstructed terraces and single-step-bunched (1×1) unreconstructed terraces. L is the faceting period and $l=L/2$ is the facet width. (c) Final stable morphology, formed below the second transition (a single-to-double step transition within the facets formed in the first transition step), consisting of (7×7) terraces and stable double-step-bunched facets with $\phi=12.7°$. θ is the surface miscut angle.

a periodic stress-domain structure [1,2], consisting of alternating (7×7) terraces and step-bunched (1×1) terraces (facets) (Fig. 3b). The change of total surface free energy per unit area is

$$\Delta F(T,L,l) = \Delta f(T)(1-\frac{l}{L}) - \frac{c}{L}\ln[\frac{L}{\pi a}\sin(\frac{l}{L}\pi)] + \frac{2\gamma}{L}, \qquad (1)$$

where T is the temperature, L is the periodicity, l is the facet width, $\Delta f(T) = f_{(7\times7)} - f_{(1\times1)}$ is the energy difference of the reconstructed (7×7) and the unreconstructed (1×1) terraces, c is a constant related to the difference of surface stress between the (7×7) terrace and the step-bunched facet and elastic constants, γ is the energy cost forming the edge of the facet, and a is a cutoff constant on the order of the (7×7) unit-cell lattice constant.

Minimization of $\Delta F(T,L,l)$ with respect to both L and l leads to the following conditions:

$$\Delta f(T) = -\frac{\pi c}{L}\cot(\frac{l}{L}\pi), \qquad (2)$$

and $$L = \pi a_0[\sin(\frac{l}{L}\pi)]^{-1}, \qquad (3)$$

where $a_0 = a\exp(1+\frac{2\gamma}{c})$. Equations (2) and (3) define the conditions and the length scales of the stress domains. $\Delta f(T)$ decreases with decreasing temperature; it is positive above the (1×1)-to-(7×7) thermodynamic phase transition temperature and negative below. The elastic relaxation drives the individual (7×7) terrace of finite size to nucleate at a temperature where $\Delta f(T) = c/(L-l)$ [3], above the (1×1)-to-(7×7) thermodynamic phase transition temperature where $\Delta f(T)$ vanishes. The ordering occurs at the lower temperatures defined by Eqn. (2). Interestingly, the ordering actually starts at a temperature coincides with the thermodynamic phase transition temperature, forming a domain structure with an optimal half-filling domain

population, $l = L/2$ [1]. Correspondingly, from Eqn. (3), the domain adopts a periodicity of $L_0 = \pi a_0$, independent of miscut angle. Using a surface stress value of 0.1855 eV/Å^2 [11] and –0.039 eV/Å^2 [12] for the (7×7) and the (1×1) terrace, respectively, and a = 47 Å, we estimate, from the observed periodicity (L_0 = 635 Å), a facet edge energy to be ~ 0.0035 eV/Å. This should be compared with the step free energy difference of ~ 0.004 eV/Å [4] between a step on the $[\bar{2}11]$ azimuth in the (7×7) and the (1×1) phase, which is determined independently from step meandering measurements, because the facet edge is formed by converting a (1×1) step into a (7×7) step. The good agreement provides a quantitative support of the proposed model.

As the temperature is lowered further, $\Delta f(T)$ becomes negative and continues to decrease. Consequently, the (7×7) terraces in every period increase their sizes, decreasing l while maintaining the periodicity L_0, to reduce the surface energy at the expense of the elastic energy. It proceeds until the most favorable facet structure is formed with a facet angle of 12.7°, as shown in Fig. 3c. Because the same facet structure is formed in all surface patterns, it is mandatory for the facet width to increase linearly with increasing miscut angle when the periodicity is fixed. The second transition step of forming the stable facet must be accompanied by a transition from single to double bi-layer step, as indicated by the final double-step-height of 6.3 Å. The single-to-double step transition is also confirmed by an independent check on step density. The total number of steps in the final patterns with ordered facets is consistently found to be half of that in the original surface.

Several additional evidence support the conclusion that there are two steps involved in the faceting transition, which define respectively two separate length scales, the periodicity and facet width of the ordered patterns. If the final patterns were formed directly without the first step, energy minimization with respect to periodicity (L) under the constraint of constant facet angle ($\phi = 12.7°$) would lead to the following relationship of periodicity and miscut angle (θ), $L_0 = \pi a_0 [\sin(\frac{\tan\theta}{\tan\phi}\pi)]^{-1}$. Consequently, the periodicity would decrease drastically with increasing miscut angle in the range from 0.3° to 6°, as shown in Fig. 2. The existence of two transition steps is also supported by the following experiment. The surfaces are first slowly cooled down to an intermediate temperature (~780°C) slightly below the first ordering temperature T_2, and then rapidly quenched to low temperature. The final surfaces exhibit the same universal periodicity (resulting from the first transition step) but different facet structure. The fast quenching rate in the second stage makes the second transition, i.e., the single-to-double step transition, incomplete. The facets contain a mixture of single and double bi-layer-height steps.

Conclusion

We have demonstrated the self-organized pattern formation on vicinal Si(111) surfaces miscut toward the $[\bar{2}11]$ direction. The patterns consist of a periodic array of alternating surface structural domains of the 7×7 reconstructed terraces and high-step-density facets. Most important, the length scales of the patterns can be controlled with a universal periodicity and linearly increasing facet width with increasing miscut angle. Such well-ordered patterns may be used as templates for subsequent nanofabrication. We propose a two-step faceting transition

model to explain the self-organization process. The first step, dominated by minimization of elastic energy, defines the universal periodicity; the second, dominated by minimization of facet energy, produces a common facet structure and controls the facet width. Future real-time experiments (such as low energy electron microscopy) can be applied to reveal the detailed dynamics of such a two-step faceting transition.

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References

1. V.I. Marchenko, Sov. Phys. JEPT Lett. 33, 381 (1981); O.L. Alerhand, D. Vanderbilt, R.D. Meade, and J.D. Joannopolous, Phys. Rev. Lett. 61, 1973 (1988).
2. F.K. Men, W.E. Packard, and M.B. Webb, Phys. Rev. Lett. 61, 2469 (1988); M.B. Webb, F.K. Men, B.S. Swartzentruber, R. Kariotis, and M.G. Lagally, Surf. Sci. 242, 23 (1991).
3. R.J. Phaneuf, N.C. Bartelt, E.D. Williams, W. Swiech, and E. Bauer, Phys. Rev. Lett. 67, 2986 (1991); R.J. Phaneuf, N.C. Bartelt, E.D. Williams, Phys. Rev. Lett. 71, 2284 (1993).
4. E.D. Williams, R.J. Phaneuf, J. Wei, N.C. Bartelt, and T.L. Einstein, Surf. Sci. 294, 219 (1993).
5. H. Hibino, T. Fukuda, M. Suzuki, Y. Homma, T. Sato, M. Iwatsuki, K. Miki, and H. Tokumoto, Phys. Rev. B 47, 13027 (1993); J.-L. Lin, D.Y. Petrovykh, J. Viernow, F.K. Men, D.J. Seo, and F.J. Himpsel, J. Appl. Phys. 84, 255 (1998).
6. M. Kasu and N. Kobayashi, Appl. Phys. Lett. 62, 1262 (1993).
7. S. Rousset, F. Pourmir, J.M. Berroir, J. Klein, J. Lecoeur, P. Hecquet, and B. Salanon, Surf. Sci. 422, 33 (1999).
8. G.M. Watson, Doon Gibbs, D.M. Zehner, Mirang Yoon, and S.G.J. Mochrie, Phys. Rev. Lett. 71, 3166 (1993).
9. P.E. Wierenga, J.A. Kubby, and J.E. Griffith, Phys. Rev. Lett. 59, 2169 (1987).
10. O.L. Alerhand, A.N. Berker, J.D. Joannopoulos, D. Vanderbilt, R.J. Hamers, and J.E. Demuth, Phys. Rev. Lett. 64, 2406 (1990).
11. R.E. Martinez, W.M. Augustyniak, and J.A. Golovchenko, Phys. Rev. Lett. 64, 1035 (1990).
12. D. Vanderbilt, Phys. Rev. Lett. 59, 1456 (1987).