Terrace-Width-Induced Domain Transition on Vicinal Si(100) Studied with Microprobe Diffraction

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We have used microprobe RHEED and a convex curved substrate to study the terrace-width dependence of equilibrium surface structures on vicinal Si(001). We observe a transition from double to single domains of the 2×1 reconstruction in periodic [110] step arrays that occurs gradually over a range of miscut angle from 0° to 5°, and is independent of temperature over the measured range of 500–800 °C. Possible mechanisms for this transition and its relation to a transition in step heights are described.

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The behavior of steps on semiconducting surfaces is an issue of fundamental importance that has received a great deal of attention recently. From an applied viewpoint, the control of step density and meander is of great practical significance for the growth of compound semiconductor overlayers and reduced-dimensional devices [1,2]. From a fundamental viewpoint, the energetics of step configurations on semiconductor surfaces is a challenging problem because there exist both short-range forces that drive surface rebonding and associated reconstructions and long-range forces that cause step interactions and associated terrace domain structures via strain fields. One can also expect that entropic effects may play a significant role in step interactions at elevated temperatures. The nature of step configurations on a vicinal surface is also of central importance to the general issue of equilibrium crystal shape and associated phase transitions [3,4].

In this Letter, we introduce a novel method for the systematic investigation of vicinal surfaces using a convex curved substrate and microprobe RHEED. A given polar and azimuthal miscut angle can be selected by the position of the beam on the curved surface. In this manner, the mean values of step and kink spacings can be continuously varied, while all other conditions of the surface, such as temperature, cleaning history, residual strain, contamination, intentional adsorbate coverage, etc., are held constant. Using this method, we have observed a terrace-width-induced transition from a double-domain to a single-domain pattern of 2×1 surface reconstruction. Current models of step interactions on semiconductor surfaces fail to explain our observations even in qualitative form.

The apparatus is a custom-built UHV scanning electron microscope with a field-emission source and dark-field-imaging capability [5]. The probe size is 50 nm at 15 keV, with 10-nA beam current [6]. Statistical step configurations are determined from the angular profiles of RHEED patterns, using well-established methods [7,8]. RHEED patterns are recorded with a vidicon system, digitized, and stored for subsequent analysis. The curved substrate is prepared from a 0.3-mm-thick (001) wafer by grinding a circular track 80 μm deep and 3 mm in diameter with a specially shaped wheel on a dimple grinder. This surface presents a continuous range of local polar miscut angles of $\pm 9°$ and a full range of azimuthal miscut angles. The sample is precleaned with hot acids followed by a light HF dip, then further cleaned in UHV by flashing to 1200 °C, followed by slow cooling to room temperature. This results in a 2×1 RHEED pattern with no sign of other reconstructions or SiC transmission spots. We verify that the curved surface is microscopically smooth (locally flat) by comparing diffraction from the bottom of the groove (maximum material removed; local miscut of zero) with that from a flat portion of the surface (no material removed). The width and intensity of diffracted beams from these regions are identical, showing that any surface irregularities from grinding or step pinning during cleaning are negligible.

On vicinal Si(100), single-layer (SL) steps of height 1.36 Å with edges along {110} separate terraces that support 1×2 and 2×1 reconstructions consisting of rows of close-packed dimers rotated by 90° between adjacent terraces. Each type of terrace can be monitored separately, since they generate different $\frac{1}{2}$-order diffraction beams. Because of the tetrahedral bonding of bulk silicon, the SL step edges on each type of terrace are inequivalent, and are labeled S_A and S_B according to whether the dimer rows on the upper terrace adjoining the step are parallel or perpendicular to the step edge, respectively [9]. Two types of double-layer (DL) steps of height 2.72 Å also occur, and are labeled similarly as D_A and D_B.

For our measurements, it is desired that the sample miscut azimuth be exactly along [110], since this will minimize the density of forced kinks. This is accomplished by finding a path along the surface for which the "dimer correlation" switches from R^+ to R^- [10]. We have found that this switch occurs in a fourfold pattern that outlines the {110} azimuths [11]. At the same time, the beam azimuth is positioned exactly along [010], which is a mirror symmetry plane for bulk silicon. This allows us to directly compare the integrated intensities and angular profiles of the 2×1 and 1×2 spots, since the effective structure factor for the two types of terrace and the scattering parameters for each beam are equivalent by symmetry [12]. RHEED patterns are recorded for a series of polar miscut angles by translating the sample in equal steps under the beam. The miscut angle is determined from the splitting of the diffraction spots. Angles less than $\sim 1°$ are determined by extrapolation, using the

macroscopic shape of the curved surface as measured with a stylus profilometer.

In Fig. 1 we show the integrated spot intensities for the $(\frac{1}{2},0)$ beam (dimer rows parallel to the step down edge), and $(0,\frac{1}{2})$ beam (dimer rows perpendicular to the step down edge) as a function of miscut angle along [110] for sample temperatures of 25 °C and 800 °C. We believe the step configuration measured at room temperature is characteristic of thermal equilibrium at a freeze-in temperature of 400–500 °C, where the step motions cease during the process of cooling from a 900 °C anneal [13–15]. The spot intensity is derived by integrating over a rectangular region of k space corresponding to 0.1×0.5 of the 1×2 Brillouin zone and subtracting a diffuse background approximated by a linear two-dimensional fit. We see in Fig. 1 that the shapes of the transition curves for the two domain types are complementary, meaning that the sum of the two intensities is essentially independent of miscut angle. This demonstrates that the linearity of the detector, the kinematic interpretation of the spot profiles, and the method of integration are accurate. Therefore, these intensities are proportional to the population of dimers of each orientation, or equivalently, to the area of each type of terrace, since the reconstruction extends fully to the step edge [16]. We see that the fraction of the surface populated by "minority" dimers (those on the upper terrace adjoining S_A steps) decreases gradually with increasing miscut angle over a range of $\sim5°$, centered at 2.5°, while the population of "majority" dimers (those on the upper terrace adjoining S_B steps) increases in a complementary fashion [17]. The data at 800 °C are seen to overlap perfectly with the room-temperature data after division by a Debye-Waller factor of $e^{-2M}\sim2.5$.

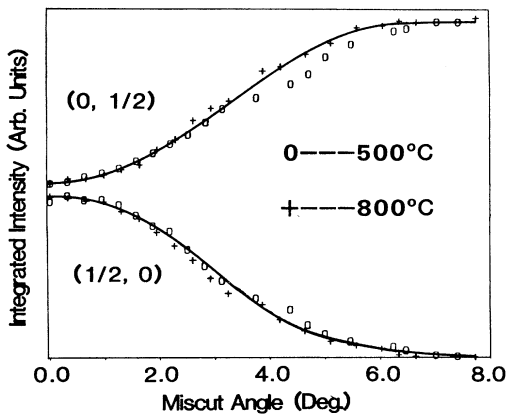

FIG. 1. Integrated intensities of $(\frac{1}{2},0)$ and $(0,\frac{1}{2})$ spots vs miscut angle along [110] for sample temperatures of 800 °C and 25 °C. The latter represents the equilibrium surface at a "freeze-in" temperature of 500 °C. Spot intensities are proportional to 1×2 and 2×1 dimer populations. The phase transition is characterized by the imbalance of dimer populations vs miscut angle. Estimated systematic uncertainties are $\pm\frac{1}{2}°$ and $\pm15\%$ of intensities.

Details of the step configuration may be deduced from the angular profiles of the diffraction spots. The Fourier transform of a line shape measured with a slit detector yields the distribution function for lengths of dimer rows on a given terrace running in the direction of the line scan [18]. We have used a point detector, but the qualitative features of the line scan are not affected. We show the majority-terrace line shape measured across the step edge, and the minority-terrace line shape measured along the nominal step edge. Looking first at the majority-terrace line shape, we see a well-defined splitting that changes continuously over the entire range of miscut angles. This corresponds to a majority-terrace-width distribution function that is narrowly peaked near $L_{\rm maj}=2h/\tan\theta$, where $2h$ is the height of a biatomic step, and θ is the macroscopic miscut angle. In the minority-terrace line shape, we find two components: a sharp central spike and a broad pedestal. This line shape corresponds to a mixture of very long dimer rows (longer than the instrumental limit of ~200 Å) and short dimer rows of length ~30 Å. The fraction of long dimer rows on the minority terrace decreases gradually with increasing miscut angle, becoming essentially zero at 4°. We conclude that the terrace domain structure changes with polar miscut angle as shown in the figure. The meander of the rough S_B-type step essentially covers the minority terrace for miscut angles larger than a few degrees. This agrees with STM observations for 2° or 3° miscut angles [19].

To interpret our results, it is first useful to briefly summarize a related transition in step heights which has received considerable attention recently. The step configuration is indirectly related to the terrace domain structure since a double-domain structure must contain SL steps, while a single-domain structure must contain exclusively one type of DL step. From electronic structure calculations of short-range, step-rebonding effects, it is known that the surface energy of a D_B step array is lower than that of an alternating S_A+S_B step array for a given polar miscut angle, and the energy of a D_A step array is larger than either of the others [9]. Long-range forces also play a role. Thus, the surface reconstruction generates a compressive (tensile) stress along (across) the dimer bond direction, with associated strain fields extending into the bulk [20,21]. SL steps are then energetically favorable, since the extent of the strain field (hence, the total elastic energy) is reduced by the formation of stress domains whose boundaries are the step edges [22]. Quantitative calculations of this effect for the Si(100) surface have predicted a striped domain size of several thousand Å [21] or several hundred Å [23] depending on the energy cost of the domain boundaries (step edges), which is not well known. More recent models include thermal effects, which cause reduction of the free energy of SL steps due to the configurational entropy associated with thermally excited kinks [24,25].

In discussing our results, we focus first on long-range effects. For miscut angles less than about 2°, it is known

from STM studies that the surface contains only SL steps [16]. From Fig. 1, however, it is clear that the domain transition begins at arbitrarily small miscut angles. We exclude the possibility of current-induced effects [26] because we obtain essentially the same transition curve for {110} step arrays running with, against, and across the direction of the heating current. The terrace-width asymmetry in this region is readily understood to result from the force dipole created by the single row of minority dimers at the "rebonded" S_B-type step. Model calculations that include this force indeed show that the minimum-energy configuration has unequal terrace widths [27].

Next we discuss the short-range effects. In Fig. 1 we see a gradual increase in terrace asymmetry, continuing from 2° up to 5° or 6°. Theoretical models predict a first-order phase transition from SL to DL steps within this range of miscut angles. One cannot sense steps directly using diffraction, only the statistical size and shape of the terraces between steps. We can, however, directly compare our data with theory by comparing the terrace asymmetry implied in the latter. Current models assume nominally straight, equally spaced steps for two phases containing SL and DL steps, respectively [23,24]. This would result in a double-domain structure with equal domain populations (50%) for all angles below a critical angle, and a single-domain structure for all angles above a critical angle, with a sharp transition between these phases. This is in serious disagreement with our data, which show a very broad transition. STM results are in nominal agreement with our data, although it is difficult to compare experiments done with multiple substrates and from different groups [24]. Furthermore, previous STM results have been presented in terms of DL versus SL steps, rather than terrace widths. The broadness of the transition could result from a phase coexistence [28]; however, our data do not support this hypothesis. From Fig. 2, we see that the separation of majority terraces changes continuously with miscut angle, with no preferred separation or associated faceting. We conclude that all surface misorientations in the [110] zone are present in the equilibrium crystal shape. From the line-shape data, we see that the widths of the minority terraces are broadly distributed for any given miscut angle. We infer that the SL and DL steps probably do coexist, but that this occurs along a given S_A-type step, as shown in Fig. 2. In the formal sense of phase-transition theory, the nature of the domain transition is somewhat unclear, since it is not obvious what constitutes a given phase, nor what the appropriate thermodynamic variables and order parameter should be [4]. It is possible that with appropriate parametrization, this transition would be continuous, similar to a step wetting transition described by Chui and Weeks [29].

Last, we describe the temperature behavior of the domain transition. From the temperature independence of the terrace-asymmetry curves we infer that the free energy of the step arrays, even at 800 °C, is dominated by

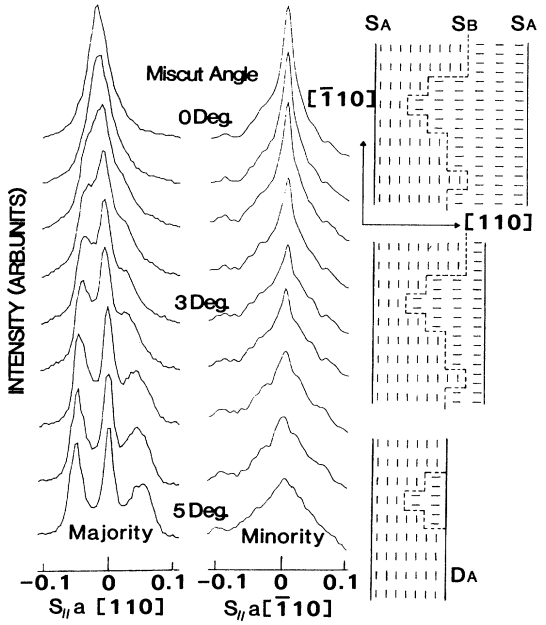

FIG. 2. Angular profiles of the majority and minority $\frac{1}{2}$-order spots measured across and along the nominal step edge, respectively, for a range of macroscopic miscut angles. The horizontal axes are plotted in units of $\mathbf{S}_{\parallel}\cdot\mathbf{a}$, where $\mathbf{S}_{\parallel}$ is the momentum transfer along the surface and $\mathbf{a}$ is the 1×1 surface lattice constant. The terrace domain structure inferred from the line shapes is shown on the right, for miscut angles of 0°, 3°, and 5°. The S_A steps are straight and regularly spaced, while the S_B steps are rough. For miscut angles larger than a few degrees, the S_B step meander covers the minority terrace, and single-layer and double-layer steps coexist along a given S_A step.

mechanical, not entropic components. This is contrary to prevailing models of step energetics on Si(100), in which configurational entropy dominates the free energy of the SL steps at temperatures above 500 °C [23–25,30].

We speculate that the dominant effect in the domain transition is a strain-mediated interaction that induces a large-scale meander of the S_B step edge, which may be described as a negative line tension. Since the step meander covers the minority terrace, there results a two-dimensional, checkerboard domain structure, albeit considerably disordered. Clearly, elastic energy may be gained by the formation of domains in the direction parallel to the step edges. In this picture, the populations of SL and DL steps and the configurational entropy associated with step meander are only incidental to the energetics of the problem. A quantitative calculation of these effects would require a fully two-dimensional geometry for the surface strain field, as well as accurate values for the step and kink energies associated with the terrace domain boundaries.

In summary, we have introduced a method for isolating the terrace-width dependence of step interactions using a curved substrate and microprobe RHEED. We have ob-

served a temperature-independent, continuous transition from double domains to single domains of 2×1 surface reconstruction induced by decreasing terrace width. Current theoretical models fail to explain qualitative features of this transition. We speculate that a dominant effect is a negative line tension for step meander, caused by stress domains in the direction parallel to the step edges.

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