Disordering of the (3×1) Reconstruction on Si(113) and the Chiral Three-State Potts Model

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We have used high-resolution low-energy electron diffraction to study the disordering of the (3×1) reconstruction on Si(113). We find that the reconstruction disorders via a continuous transition at approximately 850 K. Above the transition temperature the positions of the superlattice diffraction beams shift away from their commensurate positions. The shift is proportional to the broadening of the beams, as expected from basic scaling arguments for a system in the chiral three-state Potts-model universality class. Also consistent with numerical studies of this class is the observation of critical exponents α, β, γ, and ν approximately equal to those of the pure three-state Potts model.

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Based on Landau theory, the continuous disordering transitions of commensurately ordered surface phases were believed to fall into just four two-dimensional universality classes.[1] Systems within a universality class share certain properties near the transition, such as critical exponents. Huse and Fisher[2,3] pointed out that this classification scheme is incomplete: There is a distinctive universality class associated with the chiral Potts model.[4] In this Letter we present measurements on the disordering of the (3×1) reconstruction[5] on Si(113) which suggest it is a physical realization of the chiral three-state Potts model universality class. The (3×1) reconstruction on Si(113) has been previously studied with low-energy electron diffraction (LEED),[6-10] transmission electron microscopy,[11] and scanning tunneling microscopy,[12] but its atomic structure is still not completely determined.[7,8] A frequently observed (3×2) reconstruction[6,7,9] is metastable.[10]

Figure 1 shows schematically the (3×1) unit cell on Si(113). The ground state of the (3×1) reconstruction is threefold degenerate; domains in two of these ground states are drawn in Fig. 1. A peculiar property of the (3×1) reconstruction, evident from Fig. 1, is that the structure of domain walls perpendicular to the period-tripling direction depends on the "handedness" of the walls: If one labels the three domains A, B, and C, then $A|B$ walls are different from $B|A$ walls, for example.[2,3] This distinction does not appear in the three-state Potts model. Various theories[2,3,13] have shown that this "uniaxial chirality," which distinguishes the (3×1) from the three-state Potts model, is a relevant perturbation, in a renormalization-group sense, and thus should change the universality class.

Huse and Fisher[3] pointed out that a signature of chirality in a diffraction experiment is a shift of the diffraction peaks away from their commensurate positions as these peaks broaden above T_c. If chirality is important in determining the universality class, then the product of the shift and the inverse width of the diffraction beams should approach a constant as the transition is approached from above. Huse and Fisher predicted that this constant is a universal number characteristic of the chiral Potts model.

In our experiment an n-type Si(113) (14–16 Ω cm) wafer was used. The sample was cleaned by first outgassing at 870 K for several hours, followed by annealing to 1520 K for one minute while keeping the pressure below 5×10^{-10} Torr. The sample was cooled quickly to 1170 K and then slowly (0.2 K/sec) to room temperature. This procedure has been shown to yield clean and well-ordered Si surfaces.[14] The sample temperature was monitored by a W-(5% Re)/W-(26% Re) thermocouple mounted on the edge of the sample. The thermocouple was calibrated in the temperature range from 1020 to 1520 K against a disappearing filament pyrometer. A feedback controller stabilized the temperature to ±0.5 K. The relative uncertainty for temperature was about 2 K, while the absolute uncertainty was 60 K in the transition region (~900 K) where extrapolation of the temperature calibration was required. Diffraction profiles were measured with a high-resolution LEED system with a 500-Å transfer width.[15] Integrated intensities were measured in a different vacuum system with a different sample heater using a commercial LEED system. The LEED data were recorded with a computerized video-camera system.

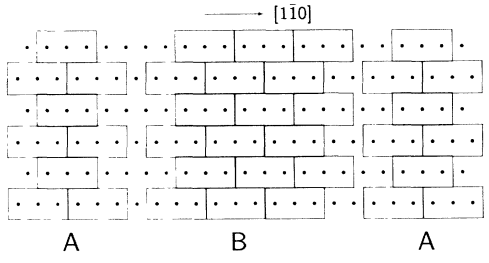

FIG. 1. A schematic picture of the surface of Si(113). The solid circles represent the position of the upper-most layer of atoms of the bulk truncated surface; the boxes represent (3×1) unit cells. Interfaces between A and B domains are shown. Notice that $A|B$ walls are distinct from $B|A$ walls.

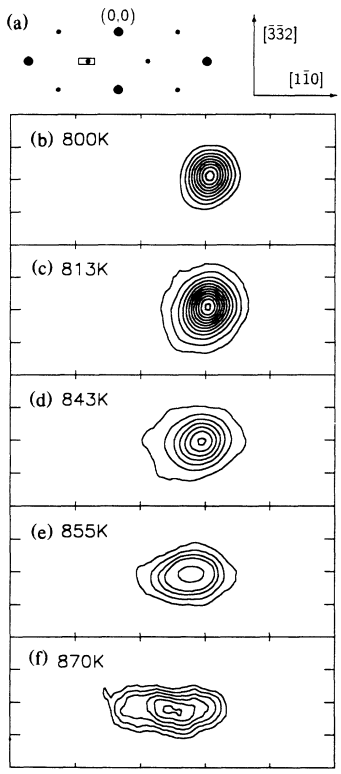

FIG. 2. Contour plots of the diffracted intensity around a third-order beam as the temperature is raised through the critical temperature (~844 K). (a) The location of the (3×1) beams in reciprocal space: The larger solid circles represent the integer-order beams. The dimensions of each of the contour plots is $\Delta k_\perp \times \Delta k_\parallel = (0.25 \times 0.09) a^{*2}$, where a^* is the third-order beam spacing in the [1 1̄0] direction. (b) The sharp third-order beam characteristic of the (3×1) at low temperature. (c)–(f) A sequence of higher temperatures: As the profiles broaden, they shift.

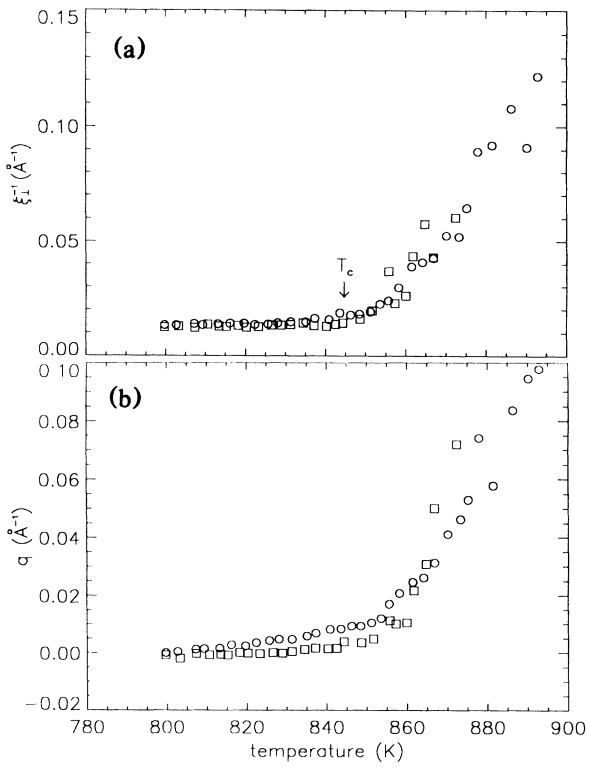

FIG. 3. (a) The inverse correlation length ($\xi_\perp^{-1}$) and (b) beam shift (q) as functions of temperature. Circles correspond to cooling and squares to heating.

Figure 2 shows a series of contour plots of the diffracted intensity around a third-order beam as the sample is heated through the disordering transition. Both the shift and broadening in the direction perpendicular to the domain walls of Fig. 1 (i.e., the [1 1̄0] direction; see Fig. 2(a)) are evident above the critical temperature. The direction and magnitude of the shift are approximately independent of the third-order beam index, thus the shift is neither a vagary of multiple scattering nor caused by surface steps.[16] To be quantitative, we fitted the third-order profiles to a Lorentzian form about the commensurate position ($\mathbf{k} \equiv 0$):

$$I(\mathbf{k},T) = \frac{\chi}{1+\xi_\perp^2 (k_\perp - q)^2 + \xi_\parallel^2 k_\parallel^2} . \quad (1)$$

Figures 3(a) and 3(b) show the beam width in the [1 1̄0] direction $\xi_\perp^{-1}(T)$ and the shift $q(T)$, respectively, as the sample is cooled and then reheated through the transition. The temperature where the broadening begins is the same as the temperature of the onset of the shift. The magnitude of q for the highest temperature measured is about 8% of the third-order beam spacing in the [1 1̄0] direction. The smallness of the difference between heating and cooling demonstrates that the transition is reversible.

Figure 4 shows $q\xi_\perp$ as a function of reduced temperature, $t \equiv (T - T_c)/T_c$. As the temperature approaches T_c (~844 K) from above, $q\xi_\perp$ approaches a constant value of about a half. Also plotted in Fig. 4 is the result of a Monte Carlo simulation of the disordering of the (3×1) phase in the generalized hard hexagon model.[17] As the temperature approaches T_c in the simulation, $q\xi_\perp$ approaches a value similar to what we observe and consistent with the prediction that this number is universal.[2,3] If there were an incommensurate phase[13] intervening between the commensurate (3×1) and disordered phase, one would expect $q\xi_\perp$ to *diverge* as the incommensurate phase was approached from above. The variation of $q\xi_\perp$ is much smaller than that observed in x-ray studies of the incommensuration of ($\sqrt{3}\times\sqrt{3}$) Kr overlayers on graphite.[18]

To study further the critical properties of the transition, we used the temperature dependence of the integrated intensity of a superlattice beam to extract the specific-heat exponent α. The integrated intensity is ex-

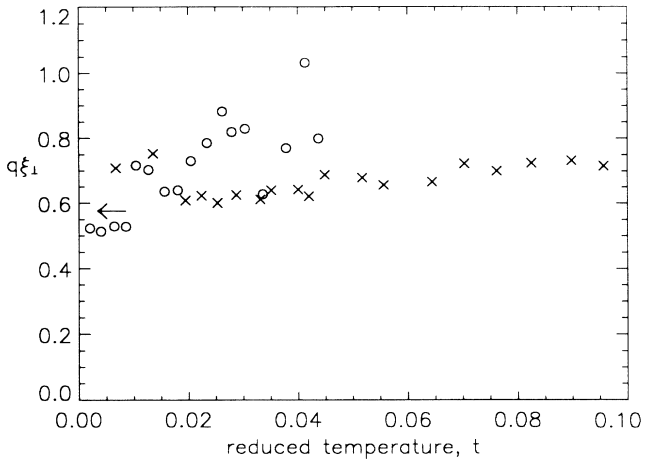

FIG. 4. A comparison of the values of $q_\perp\xi$ obtained from the data of Fig. 3 (circles) with the two-dimensional lattice-gas simulations of Ref. 17 (crosses). (The value of $q_\perp\xi$ in the limit of small t for the one-dimensional chiral Potts model is marked by the arrow.)

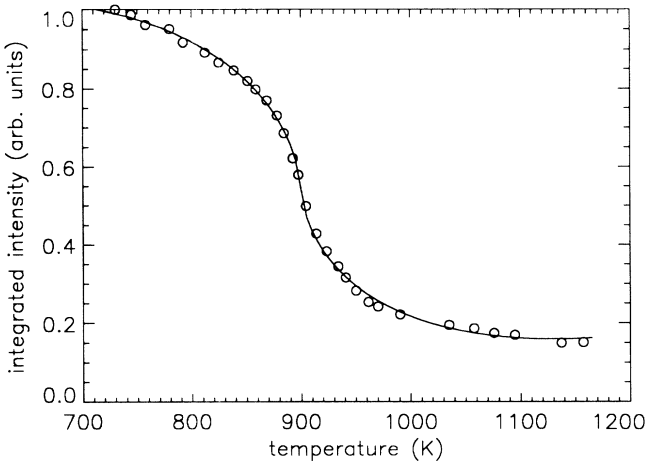

FIG. 5. Integrated intensities, obtained using a commercial LEED system, as a function of temperature. The integration radius is about 8% of the third-order beam spacing in the $[1\bar{1}0]$ direction. The solid line is the least-squares fit by Eq. (2), resulting in $\alpha = 0.32 \pm 0.06$.

pected to have the following form:[19]

$$I(T) \sim A \mp B_\pm |t|^{1-\alpha} - Ct + \cdots . \quad (2)$$

All four coefficients are positive, and the upper and lower sign refer to above and below T_c, respectively. Figure 5 shows the temperature dependence of the intensities. The inflection point in the integrated intensity plot gives an estimate of the critical temperature.[19] Depending on the integration radius and the temperature range of the fitted data, the estimated α varies from 0.26 to 0.64. Such variations are similar to numerical simulations of models in the three-state Potts-model universality class.[19] If we choose the same integration radius and fitting range which in the numerical simulations gave optimal agreement with the theoretical value of $\alpha = \frac{1}{3}$, then $T_c = 900$ K and $\alpha = 0.32 \pm 0.06$; the ± 0.06 is the statistical uncertainty of the fit. This fit is shown by the solid line in Fig. 5. Thus, even though chirality has caused the third-order beam to shift away from its third-order position by a considerable amount, the exponent is still indistinguishable from the three-state Potts model to within the experimental error. This critical behavior is consistent with previous numerical studies of the three-state chiral Potts model.[20]

Using the parameters from the fits of the measured profiles by Eq. (1), we also fitted[21] the beam intensities below and above the critical temperature and the correlation length to the forms $I(0,T) \propto -t^{2\beta}$, $\chi \propto t^{-\gamma}$, and $\xi_\perp \propto t^{-\nu}$. We find $T_c = 844$ K, $\beta = 0.11 \pm 0.04$, $\gamma = 1.03 \pm 0.19$, and $\nu = 0.99 \pm 0.18$. Again, as in the numerical simulations,[17,20] these exponents are consistent with those of the three-state Potts model ($\beta = \frac{1}{9}$, $\gamma = \frac{13}{9}$, and $\nu = \frac{5}{6}$). The difference in the critical temperatures found by the low- and high-resolution measurements (which were made using different sample heaters) is within the uncertainty in our absolute temperatures.[21] Errors in the exponents due to possible nonlinearities in the temperature calibration are smaller than the statistical uncertainties quoted above.[21]

As is evident from Fig. 2, we were not able to detect any broadening in the $[\bar{3}\bar{3}2]$ direction through the transition. One would expect some broadening if the two-dimensional chiral Potts picture of disordering of the (3×1) phase were correct. The amount of broadening is less than our instrumental resolution: Correlations are always much longer in the $[1\bar{1}0]$ direction than in the $[\bar{3}\bar{3}2]$ direction. Such large anisotropies are consistent with two-dimensional chiral behavior: In the simulation of Ref. 17, the ratio of the correlation lengths in the two directions was approximately 2.5. However, in the limit of very large anisotropies, a one-dimensional picture might become a more appropriate description of the transition. As the anisotropy gets larger, the transition temperature (measured in units of interactions perpendicular to the domain walls) becomes smaller: The one-dimensional chiral Potts model has a transition into a (perfectly) ordered state at $T = 0$. As shown in an appendix in Ref. 3, in this transition $q_\perp\xi$ approaches the value of $\cot(\pi/3) \approx 0.577$, consistent with what is observed in Fig. 4.

In summary, we have shown experimentally that the (3×1) reconstruction of Si(113), which has the symmetry of the chiral three-state Potts model, demonstrates the critical behavior predicted for that universality class. The correlated shift and broadening of the diffraction beams above T_c indicate disordering by the generation of a preferred domain-wall sequence, as predicted for a

chiral system. The numerical value of the product of shift and broadening at T_c, which is predicted to be a universal constant, is measured to be approximately one-half.

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