Critical Behavior at Chiral Melting: Disordering of the Si(113)-(3×1) Reconstruction

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Results of an x-ray-scattering study of the (3×1)-to-disordered phase transformation of the Si(113) surface are reported. A continuous commensurate-solid to incommensurate-fluid transformation occurs at $T_c = 959$ K. A new two-dimensional universality class is identified and characterized. Specifically, we find that the correlation lengths in the disordered phase scale anisotropically, and that the product of the incommensurability and the correlation length along the incommensurate direction is constant.

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According to the concept of universality, the critical behavior near a continuous phase transformation depends only on general aspects of the system, such as the symmetry of the phases involved and the dimensionality of space. Thus, the phase transformations of many disparate materials fall into universality classes, each of which is delineated by critical exponents and universal constants which are independent of microscopic details. It is generally believed that two-dimensional (2D) phase transformations may be understood in terms of several well-known universality classes [1]. Therefore, the appearance of a new 2D universality class is of special interest, particularly when its critical behavior corresponds to a simple and generic statistical mechanical model, and also is unusual in its own right. The experimental characterization of such a universality class is the topic of this paper.

Transformations between the different phases of a uniaxial overlayer on a substrate allow 2D critical behavior to be studied experimentally. Thus, the disordering of a (2×1) structure is predicted to be in the universality class of the 2D Ising model, the continuous transformation from a 2D incommensurate (IC) solid to a disordered fluid phase is expected to be in the Kosterlitz-Thouless (KT) universality class, and the commensurate-solid (C-solid) to IC-solid transformation is in the Pokrovsky-Talapov (PT) universality class [1]. A transformation that is *not* at present understood theoretically, and offers the possibility of a new universality class, is the disordering of a (3×1) C solid [2–4].

Figure 1 illustrates proposed phase diagrams for the disordering of a (3×1) overlayer. At the special point on the disordering line where light and heavy domain walls have equal free energies (P), this transformation is in the three-state Potts universality class [5]. Away from P, three competing scenarios have emerged from different treatments of the three-state chiral Potts model [6], which on the basis of universality is expected to reproduce the critical behavior of a (3×1) overlayer. The first possibility, shown in Fig. 1(a), is that a direct C-solid-to-IC-fluid transformation cannot occur for any nonzero *chirality*, defined as the difference in free energy between light and heavy walls. Instead, disordering is a two step process involving a PT transformation to an IC-solid phase and, at a higher temperature, a KT transformation to a fluid phase [2]. The second possibility, sketched in Fig. 1(b) and favored by den Nijs and co-workers [1, 3], is that at small chirality there is a direct C-solid-to-IC-fluid transformation in the three-state Potts universality class. Only when the chirality differs significantly from its value at P is the C-solid-to-IC-fluid transformation replaced by successive PT and KT transformations at a Lifshitz point (L) [3].

Finally, Huse and Fisher have proposed that between P and L in Fig. 1(b), the C-solid-to-IC-fluid transformation belongs to a new *chiral melting* universality class [4]. The signature of chiral melting is that the product of the incommensurability and the correlation length approaches a constant near the critical temperature (T_c), that is $\bar{\beta} = \nu$, where $\bar{\beta}$ and ν are the critical exponents for the incommensurability and the inverse correlation length, respectively. In contrast, den Nijs expects $\bar{\beta} = 2\nu$ [1, 5]. It has not proven possible to decide definitively among these different theoretical possibilities on the basis of numerical studies [7, 8].

Here, we report the results of a high-resolution, synchrotron x-ray-scattering study of the (3×1)-commensurate-to-disordered transformation of the Si(113)

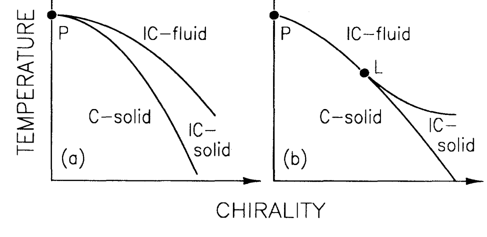

FIG. 1. Proposed phase diagrams of the disordering of a (3×1) overlayer, as described in the text.

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surface [9]. For Si(113), the reconstructed surface plays the role of the overlayer, while subsequent layers constitute the substrate. However, it is not possible to vary the chirality. Instead, it must be inferred from experiment where the disordering of Si(113) lies in the phase diagrams discussed above. Our study of Si(113) was motivated by the low-energy-electron-diffraction experiment of Yang *et al.* [9], who suggest that the Si(113)-(3×1)-to-disordered transformation provides an example of chiral melting.

We find that Si(113) undergoes a continuous transformation at $T_c = 959$ K, accompanied by critical behavior over nearly two decades of reduced temperature $[t = (T - T_c)/T_c]$ with exponents $\nu_x = 0.65 \pm 0.07$ for the inverse correlation length along the incommensurate direction (κ_x), $\nu_y = 1.06 \pm 0.07$ for the inverse correlation length along the commensurate direction (κ_y), $\bar{\beta} = 0.66 \pm 0.05$ for the incommensurability (ϵ), $\gamma = 1.56 \pm 0.13$ for the susceptibility (χ), all above T_c, and $2\beta = 0.22 \pm 0.04$ for the intensity of the commensurate peak (I_0) below T_c. Remarkably, the correlation length exponent along the incommensurate direction differs from that along the commensurate direction; that is, there is anisotropic scaling. Such behavior is very unusual and informs us that the disordering of Si(113) corresponds to a new universality class. Moreover, our results support the prediction for chiral melting that the product of the incommensurability and correlation length is a constant in the IC phase.

We studied n-type silicon wafers oriented to within 0.3° of the (113) direction, with resistivities in the range of 10–100 Ω cm. Direct current through the sample provided resistive heating, and also served as a measure of the temperature. X-ray determination of the silicon lattice constant gave a calibration, consistent with optical pyrometer measurements. The precision of the relative temperature measurement and control was 0.1 K, while the absolute accuracy is estimated to be 40 K near T_c. In addition, we estimate a temperature variation of $\sim$ 1 K across the illuminated sample area. The sample was cleaned by annealing at 1520 K for 1 min, followed by rapid cooling to 1200 K and subsequent slow cooling to temperatures of interest [9]. The UHV chamber [10], with standard surface diagnostics and integrated x-ray diffractometer, had a pressure of less than 6×10^{-10} Torr during the experiment. During sample cleaning the pressure remained below 1×10^{-8} Torr [11]. Auger electron spectroscopy indicated a clean surface, and well-ordered reconstruction peaks were consistently found.

Grazing-incidence x-ray measurements were made using the X25 wiggler beamline at the National Synchrotron Light Source. The x-ray energy was 8.8 keV. We use a rectangular coordinate system to describe the Si(113) surface. Referred to the cubic structure of Si, the surface unit cell vectors defining the x and y axes are $\mathbf{a} = (1, \bar{1}, 0)c/2$ and $\mathbf{b} = (3, 3, \bar{2})c/2$, respectively ($c = 5.43$ Å at 300 K). With this coordinate system, the unreconstructed surface is a centered rectangular structure. The corresponding reciprocal lattice vectors have magnitudes of $a^* = 1.64$ Å^{-1} and $b^* = 0.49$ Å^{-1}. The inset in Fig. 2 illustrates the 2D diffraction pattern. Squares indicate bulk truncation rods; circles represent scattering due to the reconstructed surface. At lower temperatures, the reconstruction is commensurate with the bulk, but as the temperature is raised through T_c, the peak positions shift in the directions indicated by the arrows.

Measurements were made with both high- and low-resolution spectrometer configurations. The high-resolution configuration employed a Ge(111) analyzer to define the detector acceptance. The half-height contour measured in the C phase was an ellipsoid with major and minor axes of 0.0010 Å^{-1}×0.0008 Å^{-1} in the scattering plane. In the low-resolution configuration, the in-plane angular acceptance of the detector was defined by slits to be 0.3° full width at half maximum, so that the half-height contour had dimensions 0.020 Å^{-1}×0.0012 Å^{-1}.

Figure 2 shows x-ray diffraction profiles versus q_x near (5/3,1) for several temperatures. Below T_c, the scattering profile consists of a narrow peak located at the commen-

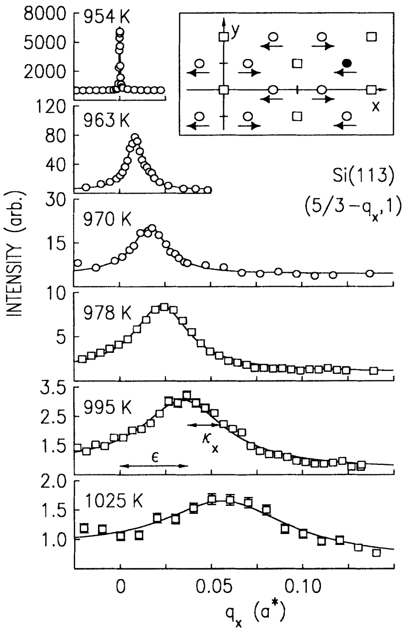

FIG. 2. Scans along the x direction through the peak of the scattering function for various temperatures. Circles correspond to the high-resolution and squares to the low-resolution diffractometer configuration. Lines are fits by the Lorentzian form described in the text. Inset is a schematic of reciprocal space: squares represent bulk scattering, while circles correspond to overlayer scattering. The filled circle is the (5/3,1) position. Arrows indicate the direction of peak shifts in the IC phase.

surate position. The full width of 0.001 Å^{-1} indicates that the (3×1) reconstruction is well ordered over several thousand angstroms. For temperatures increasing above T_c, the intensity decreases rapidly, while the peak of the scattering shifts away from the commensurate position and its width increases. In Fig. 3, the corresponding transverse profiles (versus q_y) are shown. Again, below T_c the peak is narrow, and on heating through the transformation there is an increase in the peak width. However, there is no peak shift in the y direction. No hysteresis was observed.

The peak shift reflects the formation of uniaxial walls, so that the surface layer is now incommensurate along the x direction, while remaining commensurate in the y direction. The peak broadening indicates that the translational correlation length in the IC phase is finite, that is the IC phase is a fluid. To quantify the critical behavior, we fit our profiles at each temperature above T_c to a 2D Lorentzian form for the scattering function,

$$S(q_x, q_y) = \frac{\chi}{1 + (q_x - \epsilon)^2/\kappa_x^2 + q_y^2/\kappa_y^2}, \tag{1}$$

convolved with the resolution as measured in the C phase and added to a weakly varying background. The difference between the measured wave vector and the (5/3,1) position is $\mathbf{q} = (q_x, q_y)$. Results of the fits are displayed in Figs. 2 and 3 as the solid lines. Evidently, the Lorentzian gives an excellent description of the data.

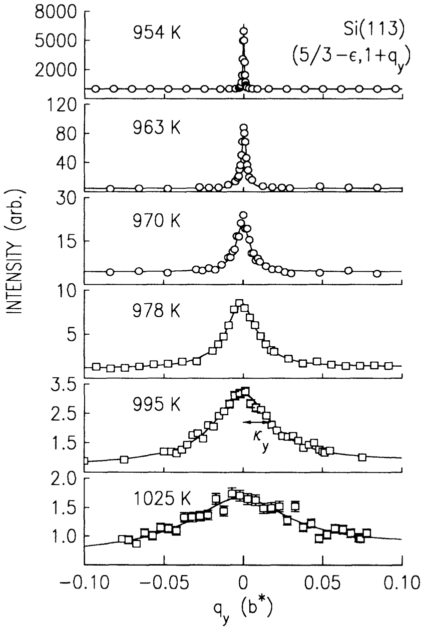

FIG. 3. Scans along the y direction through the peak of the scattering function for various temperatures. Circles correspond to the high-resolution and squares to the low-resolution configuration. Lines are fits by the Lorentzian form described in the text.

Figure 4 shows our results for the incommensurability (ϵ), the inverse correlation lengths (κ_x and κ_y), and the susceptibility (χ) versus reduced temperature. Also shown is the intensity in the C phase (I_0), which is proportional to the square of the order parameter. Each quantity shows power-law behavior over nearly two decades of reduced temperature with exponents as shown in the figure and stated above. The errors in the exponents derive primarily from our uncertainty in T_c, due to the temperature variation across the illuminated sample area. For the same reason, data analysis was restricted to reduced temperatures greater than 10^{-3}.

The results shown in Fig. 4 are striking for several reasons. First, the exponent for the correlation length along x differs from that for the correlation length along y, which is very unusual. For example, for the 2D Ising model the correlation lengths along x and y scale with the same exponent, even if the interactions are highly anisotropic. One instance in which anisotropic scaling occurs is at a PT transformation, where $\nu_x = \bar{\beta} = 1/2$ and $\nu_y = 1$ [1]. An exponent of 1/2 is excluded by our measurement of both the incommensurability (ϵ) and the inverse correlation length (κ_x), as shown by the dashed line in Fig. 4(a). Another possibility to be considered

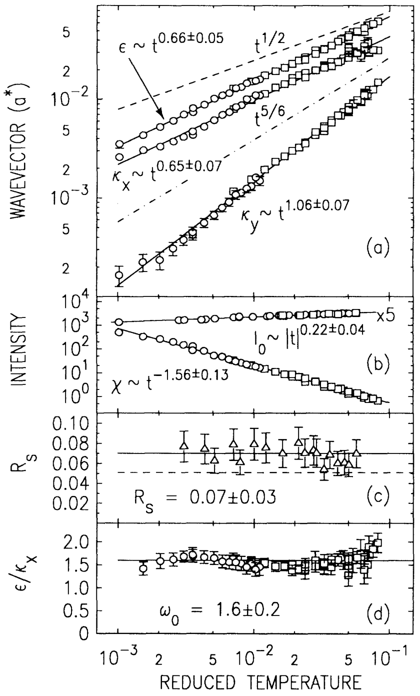

FIG. 4. (a) Incommensurability (ϵ), inverse correlation lengths (κ_x, κ_y), (b) susceptibility (χ), square of the order parameter (I_0), (c) $R_s = \chi\kappa_x\kappa_y/I_0V_r$, and (d) $w_0 = \epsilon/\kappa_x$ versus reduced temperature $t = (T - T_c)/T_c$. Circles correspond to the high-resolution and squares to the low-resolution configuration. Triangles are a combination of low- and high-resolution data from above and below T_c.

is that the exponents are those of the three-state Potts universality class, for which $\nu_x = \nu_y = 5/6$ [5]. Comparison to a 5/6 power law, shown as the dash-dotted line in Fig. 4(a), demonstrates that Si(113) disordering cannot belong to that universality class.

Second, the measured exponents are consistent with hyperscaling. Specifically, a scaling hypothesis for the correlation function near T_c gives rise to the (anisotropic) hyperscaling relation $\nu_x + \nu_y = 2 - \alpha$. In addition, a scaling form of the critical free energy leads to the relation among thermodynamic exponents $\alpha + 2\beta + \gamma = 2$. Thus, one expects $\nu_x + \nu_y - \gamma = 2\beta$, which is satisfied by the measured values. A more direct test of hyperscaling, independent of the fitted values of the critical exponents, can be made by plotting the dimensionless ratio $R_s = \chi\kappa_x\kappa_y/I_0V_r$, where V_r is the 2D resolution volume. Figure 4(c) shows that R_s is indeed independent of reduced temperature and takes the value $R_s = 0.07 \pm 0.03$. R_s may be interpreted as the ratio of the integrated intensity of the central part of the critical scattering above T_c to the order parameter scattering below T_c. In fact, according to the principle of two-scale-factor universality [12], R_s is expected to be a universal constant for each universality class. For comparison, the dashed line in Fig. 4(c) shows the value of R_s for the 2D Ising model (0.051). Finally, another universal constant, specific to the proposed chiral melting universality class, is the product of the incommensurability and the correlation length along the x direction (ϵ/κ_x), plotted in Fig. 4(d). Evidently, ϵ/κ_x is independent of reduced temperature, as expected for chiral melting, and takes the value $w_0 = 1.6 \pm 0.2$.

What are the implications of our data for the candidate phase diagrams? The existence of a direct C-solid-to-IC-fluid transformation *not* in the three-state Potts universality class indicates that the phase diagram proposed in Ref. [2] is incorrect. It follows that our data allow for two alternatives: The first, suggested by the experimental result that $\bar{\beta} = \nu_x$, is that the C-solid-to-IC-fluid transformation of Si(113) lies in the chiral melting universality class proposed by Huse and Fisher, and that its critical exponents have been determined. This interpretation finds support in finite-size-scaling calculations of a highly anisotropic chiral three-state Potts model [8]. This study reveals a C-solid-to-IC-fluid transformation exhibiting anisotropic scaling irrespective of the chirality, with exponents similar to those of Si(113). The second possibility is that the observed critical behavior is associated with the Lifshitz point (L). In this regard, based on series expansions, Howes has suggested values of ν_x and ν_y, unique to L, that are also close to those observed for Si(113) [3]. Moreover, the arguments which led Huse and Fisher to propose a chiral melting universality class seem to apply at L, leading us to expect that $\bar{\beta} = \nu_x$ there also.

Finally, this experiment demonstrates that synchrotron x-ray studies of surface phase transformations can now yield data of sufficient precision to identify and convincingly characterize new universality classes.

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