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Critical behavior of vicinal surfaces of ^{4}He

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At a temperature below the roughening temperature of a facet on a crystal, the junction between the smooth planar facet and the adjacent "rough" rounded surface should show a characteristic critical behavior, which has been calculated for several different models of surface structure. This paper presents an experimental investigation of such junction regions in hcp ^{4}He crystals next to the c facets (0001). It was found that the rough surface curves away from the plane of the facet as x^{β}, where $\beta = 1.55 \pm 0.06$. This result agree with the predictions of various terrace-step-link types of model, but disagrees with the mean-field model of Andreev.

INTRODUCTION

The experimental discovery of roughening transitions on several materials, and the intense theoretical work on these transition, which both preceded and followed the experiments, has led to several models for the microscopic structure of equilibrium crystal surfaces.[1-4] A few materials have proved themselves amenable to experimental investigations which are capable of distinguishing between the proposed models. Among them, hcp ^{4}He (Refs. 5 and 6) is the one on which the most detailed work has been carried out so far. In this paper we present experimental results pertaining to the structure of crystalline surfaces with orientations arbitrarily close to a high-symmetry direction, the so-called "vicinal surfaces," at temperatures below the uppermost roughening temperature of hcp ^{4}He.

The model which today has the strongest experimental and theoretical support for its applicability to ^{4}He surfaces is the planar *XY* model, as it is used to describe the thermal fluctuations of a faceted crystal surface through the SOS (solid-on-solid) (Refs. 7 and 8) or discrete Gaussian models. This follows from the work of Balibar and his colleagues on the growth rate of the crystals.[6] Other models include the two-dimensional (2D) Ising model, which can describe fluctuations limited to a single layer of adatoms on a facet,[9] and mean-field models, which ignore fluctuations, but which can describe the complete thermal evolution of crystal shapes.[1] In hcp ^{4}He three roughening transitions have been observed so far, on different facets.[10-12] As in most theoretical studies of phase transitions, the models make well-defined predictions about the behavior of the system close to a critical point.

Microscopic metal crystals, which like helium achieve thermal equilibrium within a reasonably short time, have also provided useful information through the study of details of their equilibrium shape. Here the agreement with one particular surface-structure model is not so clear, and of the three metals so far investigated, Pb (Ref. 13) and In (Ref. 14) appear to behave differently from Au (Ref. 15). In addition, metals have not yet shown clear evidence of roughening transitions. The work described in this paper applies the method used in these studies to the case of ^{4}He.

SHAPES OF VICINAL SURFACES

The structure of the vicinal surfaces is mirrored in the details of the transition region between a facet (macroscopically plane surface) and an adjacent rounded (atomically rough) surface. Microscopically, the vicinal surface can be pictured as a facet with more-or-less regularly spaced atomic steps of height a on it, the angle between the surface and the facet being proportional to the linear density of steps. It is useful to consider the steps as elementary excitations on the facet, with a creation energy (the step energy) and a certain energy of interaction between them. The surface free energy of the vicinal surface can then be written in a form which includes the surface free energy of the facet, the energies of the steps, their interactions, and their entropy. In general, the interaction may be repulsive (positive) or attractive (negative).

The form of the surface free energy of the vicinal surfaces has been described by several authors.[2,3,16,17] The most comprehensive description is that of Jayaprakash *et al.*,[3] which is analogous to the calculation of Pokrovskii and Talapov,[18] who discuss the structure of a monolayer deposited on a periodic substrate incommensurate with the periodicity of the monolayer itself. The steps occurring on the vicinal surface play the role of the grain boundaries separating individual commensurate regions in the latter model. Jayaprakash *et al.* investigate stepped surfaces with slope s with respect to the facet ($s=0$). The Hamiltonian is described in terms of a step-creation operator $a_j^{\dagger}$ for which the Pauli principle $[(a_j^{\dagger})^2=0]$ prevents the steps from crossing, but allows them other deformations. The solution of the resulting fermion problem gives the following dependence of free energy on the slope:

$$\gamma(s)=\gamma(0)+(a\sigma\,|\,s\,|+b\,|\,s\,|^3)/\Omega \ . \tag{1}$$

Here, Ω is the area of the unit cell on the surface, σ is the energy per unit length of a step, $b=(\pi^2/6)k_BT\exp(-\Gamma/k_BT)$, and Γ is the energy of creation of a kink in a step. This is the same form as was found by Gruber and Mullins.[17] The bs^3 term arises from the entropy of the kinks and from the effective repulsion, which prevents the steps from overlapping. There is no s^2 term. Use of the Wulff construction[4] on $\gamma(s)$ of Eq. (1) gives an equilibrium shape $y(x)$ in the vicinity of the facet edge (x_0,y_0):

$$[y(x)-y_0]/z_0=A[(x-x_0)/z_0]^{3/2}+\cdots\ , \tag{2}$$

where z_0 is the distance between the center of symmetry of the crystal and the facet and A is a constant. Inclusion of an interaction between steps with a range shorter than the distance a/s between the steps gives rise to an s^4 term in Eq. (1) and this gives a further term $D[(x-x_0)/z_0]^2$ in Eq. (2). Marchenko and Parshin[19] show that elastic interactions between the steps depend on ga^2/x^2, where g is a positive constant. To add this to (1) we replace b by $b+\pi^2g/3$, but the leading exponent $\frac{3}{2}$ in (2) is unchanged. Dipolar interactions, which can occur in metals, could lead to negative g and a discontinuity in s at the facet edge.

Jayaprakash and Saam[2] have obtained similar results for the body-centered SOS model. They derive an expansion for the profile of the vicinal surface, which meets the facet at (x_0,y_0):

$$\frac{y(x)-y_0}{z_0}=A\left[\frac{x-x_0}{z_0}\right]^{3/2}+D\left[\frac{x-x_0}{z_0}\right]^2+\cdots\ . \tag{3}$$

This formula applies to a crystal of large z_0. If the crystal is small, both the elastic energy and dependence of γ on the radius of curvature give corrections depending on $1/z_0$.[2,13] This indicates an advantage in using crystals as large as possible for an investigation. Furthermore, the coefficient A is inversely proportional to the coherence length ξ_R on the crystal surface. This is itself inversely proportional to σ, the step energy, which has a characteristic variation near T_R according to the XY model:

$$\sigma\sim\exp(-c/t^{1/2})\ , \tag{4}$$

where $t=(T_R-T)/T_R$. Thus the coefficient A approaches zero in the same manner as (4). In order to investigate the validity of the $\frac{3}{2}$ power law *per se*, it is therefore advisable to work at temperatures not too close to T_R. On the other hand, investigation of the dependence of this coefficient on T will provide additional confirmation of the model. Jayaprakash and Saam[2] have also shown that the coefficient of $[(x-x_0)/z_0]^2$ is not temperature dependent, so that the vicinal surface approaches a cylindrical or toroidal shape as T_R is approached.

In contrast, a mean-field approach to the problem by Andreev[16] employs a Landau-type expansion of the free energy near the transition, in which s is considered an order parameter. This expansion seems to have no microscopic justification. It gives a critical dependence on $t^{1/2}$, instead of Eq. (4), and an expansion for the vicinal surface, which can be written

$$\frac{y(x)-y_0}{z_0}=C\left[\frac{x-x_0}{z_0}\right]^2+\cdots\ . \tag{5}$$

This square-law dependence contrasts the $\frac{3}{2}$ power in Eq. (2), which is the signature of the square-law effective interaction between steps.

The purpose of this work is to investigate the validity of Eq. (2) or (3) experimentally in large helium crystals. The experiments of Rottman *et al.*[13] showed that the profiles of microscopic Pb crystals were consistent with the $\frac{3}{2}$ power law. However, a later analysis of the same data by Saenz and Garcia[20] suggests that the addition of a term linear in $x-x_0$ improves the fit to the experimental data, although the physical origin of this term is not clear. Metois *et al.*[15] found the transition from facet to rounded surface in Au to be discontinuous, implying either a linear $x-x_0$ term again, or attractive interactions between steps, and recently they have reported experiments in In (Ref. 14) which confirm behavior similar to Pb. In all these experiments on metals, the crystals had typical dimensions of a few micrometers, which made curvature corrections significant. Analysis of the shape of large ^{4}He crystals has recently been reported by Babkin *et al.*,[21] who found agreement with Andreev's mean field theory [Eq. (5)]. However, the crystal profiles investigated in these experiments were in a vertical plane, and the differences in gravitational potential energy between points along the crystal profile is larger than the expected difference in the surface free energy for the same points resulting from the range of crystal orientations being investigated. It thus seems possible that gravity could affect such details of the crystal profile significantly. In our experiments, the crystal profile being investigated is in a horizontal plane, so all points investigated have the same gravitational potential, and the above uncertainty does not apply.

EXPERIMENTAL PROCEDURE

The experimental system consists of a disklike cell, 11 mm diameter and 3.6 mm high. The base of the cylinder is a beryllium copper mirror, and the cell is observed from above by reflected light. Crystals with random orientations are grown in the cell by pressurizing standard-quality ^{4}He. Out of the many crystals grown, only the few with their **b** axis $(1\bar{2}10)$ within 2° of the vertical were chosen for investigation. The crystal orientation is clear from the shape of the profile and symmetry of the facets observed during growth. The above orientation gives crystals exhibiting two **c** facets (0001) and **a** facets $(10\bar{1}0)$ (Fig. 1). The crystals have lateral dimensions as large as a few millimeters, and thickness in the vertical direction approximately equal to the capillary length, 1.4 mm. In order to investigate the exponent β for surfaces vicinal to (0001) the temperature was stabilized below T_R (1.28 K) to an accuracy of 0.002 K. At such temperatures, Wolf *et al.*[6] have shown that the equilibrium time for the facet is very long. The crystals are therefore not true equilibrium crystals. However, the details that interest us are those of the vicinal surface, which should equilibrate as a rough surface, i.e., with time constants shorter than 20 s.[22] In other words, although the crystals are not in global equilibrium, we expect the part of interest to us to be in local equilibrium. We waited for equilibration times of

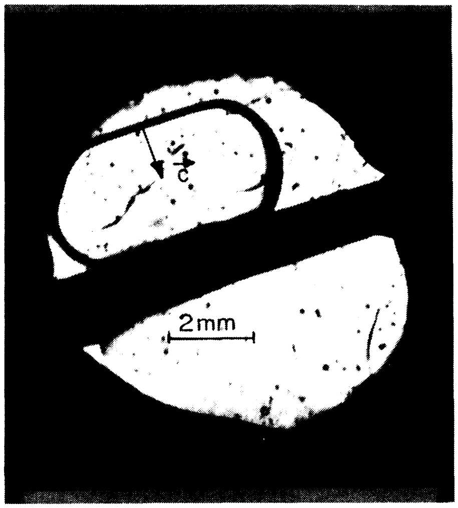

FIG. 1. Example of the profile of a crystal grown with $(1\bar{2}10)$ vertical.

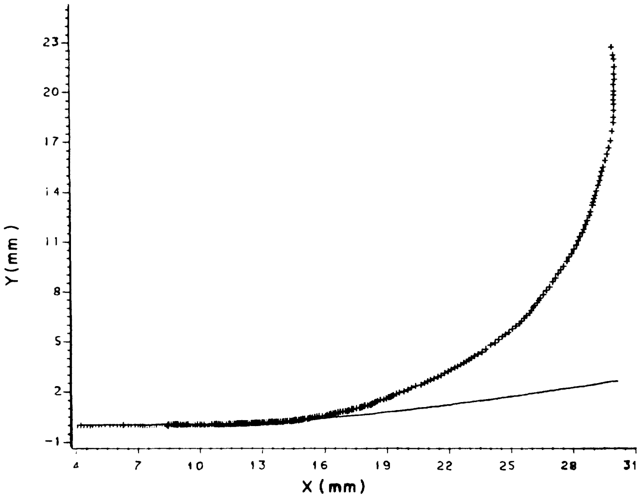

FIG. 2. A crystal profile (crosses) along the arc up to 90° from the facet, showing the fit to the vicinal region. Notice the relationship between the crystal size and the vicinal retion.

the order of 10 min. The large sizes of the nonequilibrium facets are useful in determining their orientations accurately.

The crystal profiles were photographed and magnified prints were digitized. The illumination used for the photography was from a He-Ne laser. This led to interference fringes which sometimes made the photographs difficult to analyze and, in retrospect, it might have been better to have used incoherent light. However, a sufficient number of good photographs were available. The limit of accuracy of the process was the sharpness and grain of the photographs, and was about 10 μm. About 200 points were digitized along the profile in a region containing the facet and vicinal surface up to 20° from it. Where a sufficient portion of the crystal was visible, the distance between the facets, $2z_0$, was also measured. The magnification of the photographs was about 15, but its exact value is not important in the analysis.

RESULTS

A straight line was first fitted to the part of the crystal profile corresponding to the **c** facet. Following this, the axes were rotated and translated so that the facet coincided with the x axis. Then, for an estimated location $(x_0,0)$ for the end of the facet, the curved part $(x>x_0)$, was fitted to the equation

$$\frac{y(x)}{z_0}=A\left[\frac{x-x_0}{z_0}\right]^{\beta}, \tag{6}$$

where A/z_0^{β} and β were considered free parameters. The fit was made for a range of values of x_0 around the estimated position. The final values accepted for A and β were those which gave the minimum mean-square error per point. The facet and curved region were each represented by at least 50 digitized points. The possibility of improving the fit by including the quadratic term in Eq. (3) was investigated for several photographs, D being another free parameter. Its addition made a negligible difference to the best values of A and β, but made it possible to extend the region of fit to a longer arc of the curved surface. Some further remarks on the analysis of the data are presented in the Appendix.

Figures 2 and 3 show examples of the profiles investigated. It is important to notice that what seems to be only a relatively small region close to the junction can be fitted to Eq. (6), but this is indeed in accordance with the definition of the vicinal region. Jayaprakash and Saam[2] note that this region is limited to $[(x-x_0)/z_0]\ll 1$. For the examples in Figs. 2 and 3 (in the latter the vicinal region is shown expanded), the fit was made for values of the above parameter up to 0.15. Both the digitized crystal shape and the fit according to Eq. (6) are displayed. We have analyzed 13 photographs, corresponding to four crystals at various temperatures. The values obtained for β and A are given in Table I. The coefficient A could only be obtained for crystals of which a sufficient part was visible for direct determination of the center of symmetry

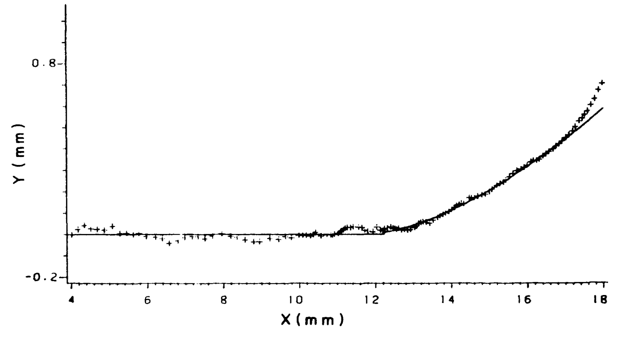

FIG. 3. An expanded view of the vicinal region of a crystal profile at 0.90 K. The crosses represent the digitized profile and the curve is the fit to Eq. (6), with $\beta=1.55$. Notice the occasional disturbance caused by fringes crossing the profile.

TABLE I. Parameters for the crystals measured.

T (K)	A	β
0.895		1.52
0.900	0.201	1.50
0.920		1.58
0.920	0.200	1.58
0.920		1.49
0.920		1.54
0.920		1.56
1.130		1.59
1.130		1.59
0.660		1.56
0.800		1.52
0.810	0.202	1.65
0.970	0.141	1.49

and the dimension z_0 to be possible. This has precluded a serious test of $A(T)$, although the general tendency for A to approach zero with increasing temperature is there.

The value of β, which best represents the data summarized above, is

$$\beta = 1.55 \pm 0.06 \ ,$$

this corresponding to vicinal surfaces up to 0.1 rad from the facet orientation. The stated limits of error represent the standard error of the set of estimates of β calculated from the 13 photographs. We have been informed that a similar study of ^{4}He crystals carried out in Paris by Gallet[23] gave a value of $\beta = 1.7 \pm 0.2$.

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APPENDIX

The most difficult point in the analysis of the data is to judge x_0, the exact point of intersection between the facet and the rounded surface. The existence of weak interference fringes on some of the photographs does not make this task any easier. It is clear from the form of Eq. (6) that if we were to choose a value of x_0 arbitrarily, we should always be able to find the value of β, which best represents the profile of the "rounded part" of the crystal, defined as $x > x_0$. In general, if x_0 is chosen too close to the center of the facet, the value of β will be larger, since the "rounded part" appears to begin more gently, and vice versa. To illustrate the interdependence of x_0 and β for one particular crystal, we show in Fig. 4 a plot, in the form of a contour map, of the mean-square error $\sum_i (y - y_i)^2/n$, where y_i is the ordinate of a digitized point and y the value according to Eq. (6). The best global value of β is that for which the mean-square error is least. One can see from the figure that an error in x_0 strongly affects the value of β, but that the optima of both can be determined. Our final value of β best represents all the crystal profiles analyzed, for which the individual values of β are tabulated in Table I.

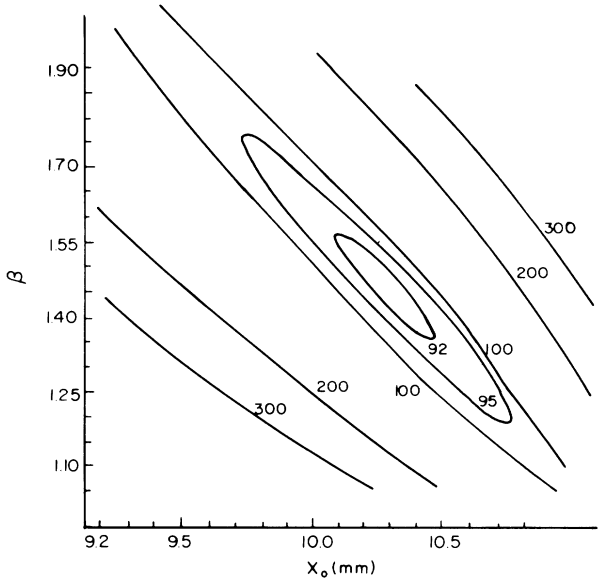

FIG. 4. Contours of the mean-square error, $10^6 \sum_i (y - y_i)^2/n$, in the x_0-β plane, for one crystal profile.

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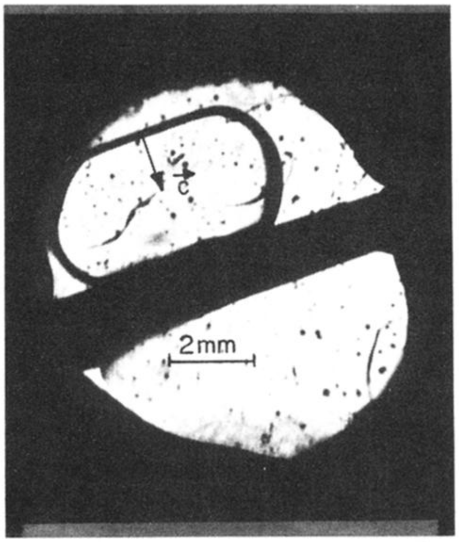

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