Transmutation of the vicinal surface exponent due to gravity

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Near the edge of a facet, vicinal surfaces curve away from the plane with exponent \( \frac{3}{5} \). With gravity normal to the facet the exponent may change to 3. Generally, there will be a crossover from 3 to \( \frac{3}{5} \) as the facet edge is approached. Comparison with experiments is made.

Vicinal surfaces are those parts of a crystalline interface that join smoothly to a facet. The limiting behavior is described by the vicinal exponent \( \theta \), defined by

\[ \delta y \sim (\delta x)^\theta, \]

where \( \delta y \) is the distance of the surface from the facet plane and \( \delta x \) is the distance from the facet edge. See Fig. 1. One reason why this number is interesting is that interface theories\(^1\) make definite predictions about \( \theta \) and measurements can be used to single out the more successful ones. Classical, mean-field theory gives \( \theta = 2 \) (at thermal equilibrium).\(^2\) Solid-on-solid (SOS) lattice-type models give \( \theta = \frac{1}{3} \).\(^3\)

Several experiments on small metallic crystals support \( \theta = \frac{1}{3} \).\(^4\) Recent, precise measurements of Carmi et al.\(^5\) for macroscopic He crystals, where the facet is oriented vertically, give \( \theta = 1.55 \pm 0.06 \) and exclude \( \theta = 2 \) by a large margin. On the other hand, older measurements of Babkin et al.\(^6\), also for macroscopic He crystals, but with the facet oriented horizontally, are inconsistent with \( \theta = \frac{1}{3} \). Although the results are not in good agreement with \( \theta = 2 \) either, \( \theta = 2 \) fits the data better than \( \theta = \frac{1}{3} \). As pointed out by Carmi, Lipson, and Polturak,\(^7\) the two He results need not be inconsistent. In a gravity-free environment, or for crystals small compared with the capillary length, horizontal and vertical facets have the same vicinal exponent. However, for large crystals in a gravitational field this need not be the case. They suggested that there are no gravitational corrections to the vicinal exponents of vertical facets, and there are such corrections for horizontal facets that tend to increase \( \theta \). One of our purposes is to show that this is indeed the case and to make predictions regarding the values \( \theta \) takes. As we shall show, horizontal facets have a crossover from \( \theta \) close to the edge to \( \theta/(2-\theta) \) away from it, provided \( \theta < 2 \). For \( \theta = 2 \), the crossover it to an exponential. The crossover region is given by \( \delta y \sim \Delta V g \), a length depending on the volume \( V \) and the gravitational acceleration \( g \). If \( \theta < 2 \), at least in two dimensions, \( \Delta \) will vanish identically if the crystal is big enough. In this case the exponent \( \theta \) is transmuted to \( \theta/(2-\theta) \) all the way to the edge. In particular, \( \theta = \frac{1}{3} \) is transmuted to 3. In three dimensions \( \Delta \) is a decreasing function of the volume. We do not know if or when it can actually vanish identically for large enough crystals. This is related to the fact that equilibrium shapes with gravity do not admit analytic solutions in three dimensions.

In this light, we have reanalyzed the data of Babkin et al.\(^6\) and found good agreement with \( \theta = 3 \). We conclude that the experiments of Carmi et al. and Babkin et al. agree and both support the SOS result of \( \theta = \frac{1}{3} \) for a gravity-free interface and disagree with the mean-field prediction of \( \theta = 2 \).

Let us introduce notation and state the basic assumptions of the theoretical framework. Denote by \( y(x) \) the crystal shape and \( p = \nabla y \) the tangent. The surface free energy is \( \sigma(p) \) and we define \( f(p) \equiv (1 + p^2)^{1/2} \sigma(p) \). In two dimensions the crystal is a film or a slab and the interface a line. \( x \) and \( p \) are then scalars. In three dimensions the interface is a surface and \( x \) and \( p \) are vectors in the plane. Let \( |p| \) be the length of \( p \) and suppose that \( p = 0 \) is the direction of the facet. Also \( L_f \) denotes the Legendre transform of \( f \), i.e., \( L_f(p) \equiv p \cdot \nabla f - f \).

Interface theories make predictions about the \( p \) dependence of the surface tension near a facet. Expressed in

FIG. 1. Schematic drawing of a crystal near a facet edge. \( \delta y \) and \( \delta x \) are the vertical and horizontal distances from the facet edge. Gravity is assumed to point along the \( y \) axis.
terms of the Legendre transform of $f$, the issue is what is
$
\mu$
in the expansion of $L_f(p)$ for $|p|$ small:
\[
L_f(p) = -f(0) + C(p) + \frac{\mu}{|p|} + \cdots,
\]
where $C(p) = p - \partial A(p)/\partial p$. If $f(p)$ has an expansion in $|p|$ then
$\mu$ must be an integer, and it must be at least 2 since the
Legendre transform eliminates the linear piece. Mean-
field theory asserts the naive expectation $\mu = 2$. SOS
models, on the other hand, predict a nontrivial cancellation
of the quadratic piece give $A(\mu = 3)$.7 The equilibrium
shape is $y = L_f(p)$, from which it follows that
\[
\theta = \frac{\mu}{(\mu-1)}.
\]
A priori, gravity could modify the analysis in two ways:
First, it could change the asymptotic behavior of $L_f(p)$.
[For example, if the coefficient of the quadratic term in
Eq. (1) is proportional to $g$, $\mu = 3$ if $g = 0$ and $\mu = 2$ if
$g \neq 0.$] Second, the relation between $\mu$ and $\theta$, Eq. (2),
needs to be modified since the equilibrium shape is no
longer given by the Wulff construction.

The basic assumption we shall make is that $\sigma(p)$ is
independent of $g$, which implies that $\mu$ is independent of $g$.
This is related to the continuum limit. Explicitly, on
height scales much smaller than the capillary length, gravity
effects are small. If this scale is still much larger
than interatomic distances, $\sigma(p)$ may be defined locally,
according to the usual statistical mechanics definition.
Thus, in the continuum limit where interatomic distances
are negligible, $\sigma(p)$ is a local property, independent of $g$.
In the rest of this paper we examine how Eq. (2) is
modified by gravity.

It is actually somewhat surprising that gravity can
transmute the vicinal exponent at all. For the vicinal
exponent [like $\sigma(p)$] appears to be a local property that is
defined on height scales that can be much smaller than
capillary lengths. This na"ive picture is false. The vicinal
exponent actually depends on global features like the
volume and actual height of the crystal. To see this, re-
call the Euler-Lagrange equation for the minimization of
the surface-free energy, and the gravitational energy sub-
ject to a volume constraint (Herring's formula8,9):
\[
\sum \frac{\partial^2 y}{\partial x_i \partial x_j} \frac{\partial^2 f}{\partial p_i \partial p_j} = \kappa(\Delta + \delta y),
\]
where $\kappa (\equiv \partial \rho / \partial g)$ is the "specific weight" of the crystal
($\Delta$ is the difference in densities between the crystal and
the surrounding medium), $\Delta \equiv (\lambda / \kappa) - h$ where $h$
is the height of the crystal, $\delta y \equiv h - y$, and $\lambda > 0$ is the
Lagrange multiplier associated with the volume
constraint. The "source term," the right-hand side of (3), is
non-negative, expressing the convexity of the equilibrium
shape. If $g = 0$, or if $\Delta > 0$ and $\delta y << \Delta$, the "source" is
essentially a fixed strictly positive constant, so the
vicinal exponents are the same. However, when $\Delta$ vanishes
(or when $\delta y$ dominates $\Delta$), the source is qualitatively
different and becomes arbitrarily weak as $\delta y$ gets small.
There is no reason why the vicinal exponent in such a
case would be as in the $g = 0$ case. Since $\Delta$ depends on
global features ($h$ and $V$) $\theta$ is not a purely local property.

The vicinal exponent of vertical surfaces is not
modified by gravity because for such surfaces $\delta y$ does not
tet to be arbitrarily small. But that of horizontal
surfaces may be modified. Before discussing this
modification quantitatively, let us give an interpretation of the
crossover scale $\Delta$.

The gravity-free equilibrium crystal surface can be
understood as the balance between the pressure difference
across the surface and the (anisotropic) surface energy.10
It is intuitively obvious that the crystal shape is convex if
the pressure is higher on the crystal side. In the presence
of gravity, the effective pressure is linear in $y$ and vanishes
at some "fiducial" height. The crystal surface clearly
does not rise above this height; otherwise it would be
cavend. $\Delta$ is precisely the difference between the fiducial
height and the maximum height of the crystal. For
crystal surfaces which are rounded at the top, Herring's for-
mula8,9 states $\Delta = \sum K_i S_i$, where $K_i$ are the principal
curvatures at the top and $S_i$ are principal radii of curva-
ture in the Wulff plot there (i.e., the eigenvalues of the
stiffness tensor11). Since $S_i$ remain fixed while the curva-
ture of the crystal is expected to decrease for increasing $g$
and $V$, $\Delta$ is expected to be a decreasing function of $V$. ($V$
may not be monotonically so for the height is known to
be nonmonotonic for liquids.)12 Thus, $\Delta$ can be made
small by enlarging the crystal. For crystals with a facet
at the top, this conclusion is not altered, though the argu-
ments need to be tightened, as Herring's formula holds
for the smooth portions of the interface.

To examine how $\theta$ changes, consider first the two-
dimensional case of a crystal supported on a flat horizon-
tal base. The Euler-Lagrange equation has a first in-
tegral9
\[
L_f(p) + \lambda y - \frac{1}{2} \kappa y^2 = \Lambda_f(p_0),
\]
where $p_0$ is the tangent of the contact angle. With a facet
at $p = 0$, subtracting $y(p)$ from $h = y(0)$ in Eq. (4) one gets
\[
\delta y (2 \Delta + \delta y) = \frac{2}{\kappa} [L_f(p) - L_f(p_0)].
\]
By (1), the right-hand side of (5) is of order $p | \mu$. Thus,
$\delta y \sim p^\mu$ if $\Delta \gg \delta$ and $\delta y \sim p^{\mu/2}$ if $\delta y \gg \Delta$, so that
\[
\theta = \begin{cases} \frac{\mu}{(\mu - 1)} & \text{if } \delta y \ll \Delta, \\
\frac{\mu}{(\mu - 2)} & \text{if } \delta y \gg \Delta. \end{cases}
\]
So, there is a crossover at a scale $\Delta$. If $\Delta = 0$, the
exponent is transmuted all the way to the edge of the facet.
In particular, if $\mu = 3$, $\theta = 3$ for $\delta y$ larger than $\Delta$ and $\frac{1}{2}$ for
$\delta y$ smaller than $\Delta$. From Ref. 9 $\lambda$ is a nonincreasing and
$h$ a nondecreasing function of $V$ so $\Delta$ is a nondecreasing
function of $V$. In fact, if $\mu > 2$, $\Delta$ vanishes for $V > V_0$
where $V_0$ is the finite critical volume whose part lying to
the right of the facet, $V_0'$ is
\[
2 \kappa V_0' = \int_{p_0}^0 \left[ 1 - \frac{2 \kappa}{\lambda^2} \frac{[L_f(p) - L_f(p_0)]^{1/2}}{f''(p)} \right] dp.
\]
The three-dimensional case is, of course, more complicated. We restrict ourselves to the case of cylindrical symmetry where the Euler-Lagrange equation is the (nonlinear) differential equation
\[ \frac{1}{kr} \frac{\partial}{\partial r} \left( r f'(r) \right) = (\Delta + \delta y) . \]
(8)

Here \( r \equiv x \mid r \), \( p \equiv \delta y / \delta r \). Let \( r_0 \) be the edge of the facet, \( f'(r) = s + \text{const} \times r^{(\mu - 1)} \) and \( \delta y \sim (\delta r)^\theta \), with \( \theta > 1 \) (\( s > 0 \) is the step-free energy). The left-hand side of (8) is, to leading orders,
\[ \frac{1}{kr_0} \left[ s + (\text{const}) (\delta r)^{(\mu - 1)(\theta - 1) - 1} + \cdots \right] . \]
(9)

There are two possibilities: Either \( r_0 \Delta = s / \kappa \), in which case \( (\mu - 1)(\theta - 1) - 1 = \theta \), or \( r_0 \Delta \neq s / \kappa \), in which case \( (\mu - 1)(\theta - 1) - 1 = 0 \). The first gives \( \theta = \mu / (\mu - 2) \) and the second gives \( \theta = \mu / (\mu - 1) \). Thus, again, depending on \( \Delta \) there is a transmutation of \( \theta \). Also, as in two dimensions in the general case, there is a crossover.

In light of the transmutation of \( 2/3 \) to 3, we took the data of Babkin et al., \(^6\) presented in their Fig. 3b, and plotted \( \sqrt{\delta} \) against \( x \). Since \( \delta \) is essentially \( p \) at these angles, \( \delta y \sim (\delta x)^\theta \) predicts that our plot should be linear, with a crossover to \( (\delta x)^{3/4} \). In Fig. 2, we see that the first two points are still on the facet while all but the last two or three lie on a straight line. We cannot quote how well a straight line fits the data, since we have neither the numerical values nor the errors. (One difficulty is that it is not obvious how many points actually lie in the asymptotic region of vicinal surfaces. As pointed out to us by Lipson, using a criterion of Jayaprakash and Saam in Ref. 3 to define the vicinal region, his analysis of the data of Babkin et al. suggests that the scale is about 2 mm, in reasonable agreement with the 2.8 mm suggested by our fit.) The overall agreement supports the claim of transmutation.

From the graph we can also get an estimate of \( \Delta \). The crossover appears to occur between the second point, which is on the facet, and the third point, which is already on the straight line. This corresponds to \( \sqrt{\delta} \) of the order of 0.3 and so to \( \phi \) of order 0.01, and \( \delta x \) of order of a fraction of a mm. This gives \( \delta y \), and \( \Delta \) of the order of a micrometer. With more data between the second and the third point, \( \Delta \) could be determined more accurately.

It is probably no coincidence that data in the crossover region are not reported. The authors of Ref. 6 pointed out the uncertainties concerning the lack of equilibrium \(^1\) for facets and their neighborhoods. Thus, even if measurements were taken in this region, it would be difficult to untangle the effects of nonequilibrium from the critical behavior of vicinal surfaces. In this sense, there is much theoretical work to be done on the difficult issue of dealing with surfaces in equilibrium joined to those not in equilibrium.

If the external force is not uniform but depends on \( y \), it is possible for the exponent to be transmuted some other way. This can be traced to the third term in Eq. (4). If its power were \( m \) instead of 2, which came from the specific form of the gravitational potential, then the transmuted exponent will be \( \theta / (m - (m - 1)) \). Nonuniform forces can be produced in centrifuges or between uneven condenser plates, for examples.

Finally, similar ideas can be used to analyze the effects of gravity on the universal jump of the curvature near the roughening transition. \(^{13,14}\)

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8C. Herring, Phys. Rev. 82, 87 (1951).


