Roughening transition in the interface between superfluid and solid ^{4}He

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The equilibrium shape and dimensions of a small crystal are presented by consideration of its anisotropic surface tension and gravitational effects. The roughening transition is discussed phenomenologically in terms of the facet stabilizing *cusp points* appearing in the surface tension. This is applied to recent experiments on the interface between superfluid and solid ^{4}He.

The equilibrium structure of a crystal surface and the existence of a surface roughening transition at a critical temperature T_R was discussed by Burton, Cabrera, and Frank.[1] The most convenient method for studying the shape of a crystal at equilibrium would be to use the surface free energy and Wulff's theorem.[2] These surface free energies of crystals $\alpha(n)$, where n is the normal to the surface, were also discussed by Herring.[3] Cabrera[4,5] studied the stability and shape of different surface orientations considering the behavior of $\alpha(n)$. In all the above references it is perfectly clear that the existence of faceting and plane singular surfaces of crystals is related to the existence of *cusp points* in the surface tension $\beta(p,q)$ referred to a stable singular surface:

$$\beta(p,q)=\alpha(n)(1+p^2+q^2)^{1/2} \ , \tag{1}$$

where the surface $Z=\zeta(x,y)$ is defined by the slopes $p=-(\delta\zeta/\delta x)$ and $q=-(\delta\zeta/\delta y)$. The shape[6] of the surface and its dimensions[5] can be obtained by minimizing the thermodynamic potential Ω, as proposed by Landau.[6]

In this Communication we present a study of the shape and dimensions of crystals in equilibrium with gas or liquid phases by using a perturbative expansion of the surface tension $\beta(p,q)$ near to a plane singular surface $p=q=0$. We discuss the roughening transition in terms of the steps surface energies[4,5,7,8] as a function of (p,q) and temperature T. We believe that this can be applied to explain recent experimental results by Balibar *et al.*,[9] Landau *et al.*,[10] and Avron *et al.*[10] that recognized the existence of the roughening transition for hcp ^{4}He.

The difference in thermodynamic potential $\Delta\Omega$ between phases neglecting tensions and strains in the crystal reads as[11]

$$\Delta\Omega=-\int_B dx\,dy\left[\int_0^{\zeta(x,y)}\Delta P\,dz + [\Delta\rho g(\tfrac{1}{2}\zeta^2-Z_0\zeta)]+\beta(p,q)\right] \ , \tag{2}$$

where ΔP and $\Delta\rho$ are the supersaturation and difference in the density between phases and g is the gravity. Now we must determine the surface $\zeta(x,y)$ by minimizing $\Delta\Omega$, which already includes the total volume of both phases constant. The minimization results in the Euler equation[5,6]

$$\Delta P_\zeta+\Delta\rho g(\zeta-Z_0)=\frac{\partial^2\beta}{\partial x\,\partial p}+\frac{\partial^2\beta}{\partial y\,\partial q} \ , \tag{3}$$

where Z_0 is a reference height and ΔP_ζ is the jump in pressure at the surface ζ that determines the shape and dimensions of the crystal. The solution to this Euler equation given by Landau[6] [Eq. (155.3)] and also in Ref. 5 is

$$\zeta(x,y)=\frac{2}{\Delta P_T}\left[\beta-p\frac{\partial\beta}{\partial p}-q\frac{\partial\beta}{\partial q}\right] \ , \tag{4}$$

with

$$x=\frac{2}{\Delta P_T}\frac{\partial\beta}{\partial p}, \quad y=\frac{2}{\Delta P_T}\frac{\partial\beta}{\partial q} \tag{5}$$

and

$$\Delta P_T=\Delta P_\zeta+\Delta\rho g(\zeta-Z_0) \ . \tag{6}$$

Then formulas (4)–(6) provide us with $\zeta(x,y)$, if β is known. We propose to expand this function in (p,q) as has been previously done.[3,5,7,8] For (p,q) small we have

$$\beta(p,q,T)=\beta_0+\beta_1|p|+\beta_2|q|+\beta_3p^2+\beta_4q^2+\beta_5p^4+\beta_6q^4+\cdots \ , \tag{7}$$

where the interpretation of the coefficients $\beta_i(i=0,\ldots,n)$, which are T dependent, is as follows: (i) $\beta_0>0$ is the surface tension of the reference singular surface; (ii) β_1 and $\beta_2>0$ is the excess surface energy corresponding to the formation of a step.[5,7,8] This is the basic cusp term which drops to zero at T_R[6,7,8]; (iii) β_3 and β_4 are interaction surface energies between steps; and (iv) β_5 and β_6 correspond to the interaction between cross-correlated steps.

For simplicity, we discuss a case in which $\beta_i=0(i>5)$, although for fitting experimental data[9,10] we will use up to $i=6$. By using formula (5)

we have

$$p \equiv \frac{\frac{1}{2}\Delta P_T x - \beta_1[2\Theta(p)-1]}{2\beta_3} \ ,$$
$$q \equiv \frac{\frac{1}{2}\Delta P_T y - \beta_2[2\Theta(q)-1]}{2\beta_4} \ , \tag{8}$$

where $\Theta(h)$ is the Heaviside function. The basic result is that for the ranges in x and y, $|(\Delta P_T/2)x| \leq \beta_1$ and $|(\Delta P_T/2)y| \leq \beta_2$, the only solution is p and $q=0$; i.e., the crystal has a stable singular facet of area $16\beta_1\beta_2/(\Delta P_T)^2$, and for x and y larger than the above ranges we have

$$0 = \zeta - \frac{2}{\Delta P_T}\left[\beta_0 - \frac{(\frac{1}{2}\Delta P_T x - \beta_1)^2}{4\beta_3} - \frac{(\frac{1}{2}\Delta P_T y - \beta_2)^2}{4\beta_4}\right] , \tag{9}$$

where it should be remembered that ΔP_T is a function of ζ. It is worth noting that around T_R, theory[7(c)] shows that β_1 or β_2 behave like $\epsilon_1 \exp(-\chi|T-T_R|^{-1/2})$ and then the singular facet surface at T_R disappears as

$$x_F = \frac{2}{\Delta P_T}\epsilon_1 \exp(-\chi|T-T_R|^{-1/2})\Theta(T_R-T) \ ,$$
$$y_F = \frac{2}{\Delta P_T}\epsilon_2 \exp(-\chi|T-T_R|^{-1/2})\Theta(T_R-T) \ , \tag{10}$$

where (x_F, y_F) are the maxima coordinates of the facet and reduce to zero as $T \to T_R$. Consequently, at T_R, there are no facets because the excess surface is zero. *This is the roughening transition.*

We apply now the above results to the experimental data[10] on solidification of hcp ^{4}He. These experiments show clearly a top singular facet surface in the meniscus $x=0$, $\zeta(y)$, at $T=0.94$ K, that has not been explained theoretically (see Landau *et al.*,[10] Fig. 1). Actually, the data referring to the singular facet have been fitted in excellent agreement with experiments for the following values of the parameters: $\Delta P_\zeta = 1.5$ erg/cm^3, $\Delta\rho = 0.017$ g/cm^2 (taken from Grilly),[12] $\beta_2 = 0.0975$ erg/cm^2, $\beta_4 = 0.08$ erg/cm^2, and $\beta_6 = 0.01$ erg/cm^2, resulting a value of $Y_F = 0.135$ cm. These calculations are drawn in Fig. 1 where

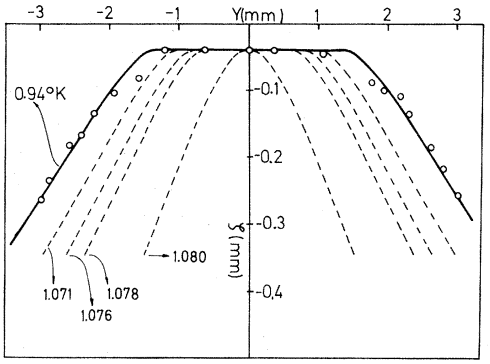

FIG. 1. Fitting of the experimental data (circles O) of (Ref. 10, Fig. 1) for $T=0.94$ K. Also, the behavior of the crystal surface is shown as $T \to T_R$.

$Z_0 = \zeta_{max} = -0.004$ cm. Also in this figure are shown curves for different temperatures as $T \to T_R \simeq 1.08$ K, as has been obtained by Avron *et al.*[10] from equilibrium holographic interferograms. From Fig. 2 of Avron *et al.*[10] we obtain a value $\chi \simeq 0.04$ K$^{1/2}$. It can be seen how the plane area is reduced as $T \to T_R$. It should be mentioned also that gravity in this case is important because ΔP_ζ is very small.

In conclusion, we have seen that the ^{4}He experiments can be understood, taking into account the anisotropy of the crystal surface tension that contains *cusp points* for $T < T_R$. The cusp points, as well as the other terms in the surface tension, should be interpreted in terms of energies and interactions between steps. Also, the roughening transition should be detected experimentally by increasing $T \to T_R$ and observing the different meniscus shapes. In fact, this kind of experiment will provide a method to determine experimentally the value of χ. Also, our results are in contradiction with those of Andreev and Parshin[13] that predict a rough surface for $T=0$ K; this is inconsistent with the existence of cusp points in the surface tension.

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[1]W. K. Burton, N. Cabrera, and F. C. Frank, Philos. Trans. R. Soc. London Ser. A 243, 299 (1951).
[2]G. Wulff, Z. Kristallogr. 34, 449 (1901).
[3]C. Herring, Phys. Rev. 82, 87 (1951).
[4]N. Cabrera, Am. Soc. Test. Mater. Proc. 340, 24 (1962).
[5]N. Cabrera, Surf. Sci. 2, 320 (1964).
[6]L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Pergamon, New York, 1980), p. 520–522.
[7](a) H. J. Leamy and G. H. Gilmer, J. Cryst. Growth 24-25, 499 (1974); (b) J. D. Weeks and G. H. Gilmer, *ibid.* 43, 385 (1978); (c) H. van Beijeren, Phys. Rev. Lett. 38, 993 (1977).

[8]H. J. Leamy, G. H. Gilmer, and K. A. Jackson, in *Surface Physics of Materials,* edited by J. M. Blakely (Academic, New York, 1975), Vol. I, pp. 121–188.
[9]S. Balibar, D. O. Edwards, and C. Laroche, Phys. Rev. Lett. 42, 782 (1979).
[10]J. Landau, S. G. Lipson, L. M. Määttänen, L. S. Balfour, and D. O. Edwards, Phys. Rev. Lett. 45, 31 (1980); J. E. Avron, L. S. Balfour, C. G. Kuper, J. Landau, S. G. Lipson, and L. S. Schulman, *ibid.* 45, 814 (1980).
[11]N. Cabrera and N. García (unpublished).
[12]E. R. Grilly, J. Low Temp. Phys. 11, 33 (1973).
[13]A. F. Andreev and Ya. Parshin, Zh. Eksp. Teor. Fiz. 75, 1511 (1978) [Sov. Phys. JETP 48, 763 (1978)].