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Roughening Transition in the ^{4}He Solid-Superfluid Interface

J. E. Avron, L. S. Balfour, C. G. Kuper, J. Landau, S. G. Lipson, and L. S. Schulman

Department of Physics, Technion–Israel Institute of Technology, Haifa, Israel

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Principal planes of the ^{4}He solid-superfluid interface are expected to undergo roughening transitions at temperatures of about 1 K. An experiment is described in which two such transitions were observed in the hcp-superfluid interface: first for the basal plane at 1.08 K and second for an orthogonal face at 0.85 K.

The roughening transition (RT) is a theoretical concept, known to apply to certain Ising models,[1] and to computer simulations of crystal growth.[2,3] In this Letter we discuss the applicability of the RT to a real physical system: the ^{4}He solid-superfluid interface. We report the observation of morphological transitions in ^{4}He crystals at 1.08 and 0.85 K which, we argue, are RT's. Parshin and his co-workers[4,5] have argued that the interface in this system is always rough, but their view is contrary to evidence presented here (and by Landau *et al*[6]). Balibar[7] has independently proposed the existence of a RT in the system.

The roughening temperature, T_R, is characterized by the vanishing of the step energy.[1,2] Above T_R the interface fluctuates macroscopically, and in the thermodynamic limit, translation invariance is restored.[8] Unfortunately, this aspect of the RT is not expected to be observable in practice. Fluctuations in ordinary macroscopic systems are microscopic; for the two-dimensional interface, the mean square amplitude fluctuation is of order $\ln N$, where N is the number of lattice points in the interface.

The interface is modeled by a two-dimensional (2D) lattice Hamiltonian with discrete, unbounded Ising spin Z: the vertical coordinate of the interface. A capillary-wave Hamiltonian[9] for the interface is

$$H = \sum_{\vec{1}} \left\{ 1 + [\nabla Z(\vec{1})]^2 \right\}^{1/2} \left[\sigma_0 \Omega + (\rho_0 \hbar^2/M^2)(\nabla \psi)^2 \right].$$

(1)

Here, ψ is the superfluid field, ρ_0 its density, ∇ the discrete gradient, Ω the area of the 2D unit cell, and $\vec{1}$ a lattice bond. The term proportional to σ_0 (a coupling constant) represents the energy density of the boundary of the solid, and the term proportional to $\hbar^2$ is the energy density of the superfluid layer near the interface.[10,11] In the lowest approximation the ψ field integrates out. Similar conclusions hold for the Villain model[12] and for other models[13] of the interface. The surface tension is the free energy of the Hamiltonian H.

The specific choice of the interface Hamiltonian is of little importance here since the Kosterlitz-Thouless roughening transition[14] is a general feature of 2D models whose Hamiltonians are invari-

ant under discrete uniform shifts $Z(\vec{1}) \to Z(\vec{1})$ $+ a$.[14,15]

The quantum aspects of solid He will be taken into account only phenomenologically; σ_0 is taken to be the measured surface tension of ^{4}He at $T = 0$.[6] The roughening temperature is given by $T_R = K\sigma_0\Omega$, where K is a constant of order unity, whose value depends on the details of the interface model (the choice of Hamiltonian), the lattice structure, etc.[16] Taking σ_0 and Ω appropriate to the ^{4}He interface[6] gives $T_R \simeq 1$ K.

Despite the difficulty of calculating T_R, we can estimate *relative* values of T_R on different planes and hence establish a *hierarchical ordering* of plane orientation according to roughening temperature. To accomplish this we identify the ^{4}He interface with the interface of a 3D Ising model with Slater-Kirkwood[17] pair interaction and $\pm$ boundary conditions. This is reasonable only below 1 K, where ^{4}He, like the Ising model, has essentially no latent heat.[18,19] To estimate T_R we use a bound of van Beijern as if it were an equality. Van Beijern proved that for a given crystal orientation $T_R \geq T_c$, where T_c is the critical temperature of the 2D Ising model obtained by considering the restriction of the 3D Ising model to the plane of the interface. This reduces the problem to that of computing T_c for certain 2D Ising models, which represent sections of the 3D structure and therefore have different lattices and interatomic couplings.

We have estimated T_c for these 2D models with the Slater-Kirkwood interaction both by mean-field theory[21] and by reduction to soluble models.[22] The results are summarized in Table I, showing that the basal plane has the largest T_R.

The roughening transition implies[23] that faces which are flat below T_R will become curved (atom-

ically rough) above it.[24] This can happen with an arbitrarily small change in $\sigma(\theta,\varphi)$ that smooths a cusp in the Wulff diagram.[24]

We have recently been investigating[6] experimentally the surface tension at the solid-superfluid helium interface. In particular, we found that σ is independent of T within a single crystallographic phase, and that large equilibrium crystals have apparently more facets at low temperatures than at high temperatures.[25] Evidence for a RT, and even for a heirarchy of such transitions, comes from observing the shape of a small[26] crystal as it was very slowly warmed up. The size of the crystal was comparable with the capillary length, 1.4 mm. A complete sequence of holographic interferograms[27] was photographed,
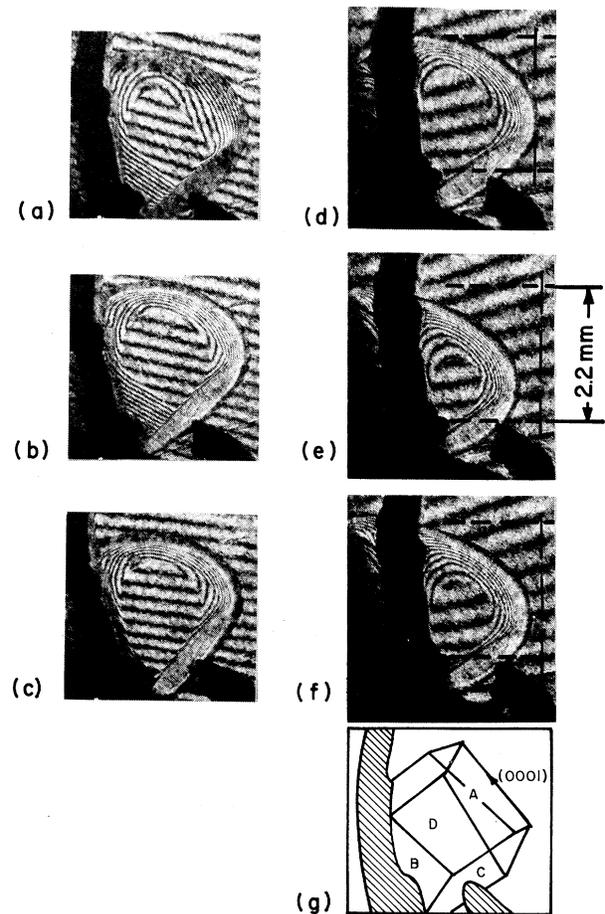

(a) (b) (c) (d) (e) (f) (g)

FIG. 1. Equilibrium holographic interferograms of the ^{4}He crystal at constant volume: (a) at 0.6 K; (b) at 0.8 K below the lower transition; (c) at 0.9 K above the lower transition; (d) at 1.06 K before the upper transition; (e) at 1.09 K after the upper transition; (f) at 1.06 K after the reversal of (d) $\to$ (e); (g) schematic of the planes in (a) (Ref. 28).

TABLE I. Estimates of the roughening temperature for various crystal planes. The temperature scale was chosen so that the highest T_R is unity. The mean-field values were obtained by summing the Slater-Kirkwood potential in the given plane. The nearest-neighbors estimate follows from nearest-neighbors soluble models with interaction fitted to the Slater-Kirkwood potential.

Miller indices	T_R, arbitrary units	
	Mean field	Nearest neighbors
(0001)	1	1
(11$\bar{2}$0)	0.5	0.5–1
(10$\bar{1}$0)	0.35	0.2
(10$\bar{1}$1)	0.34	0.16

of which several significant prints are assembled in Fig. 1. At the lowest temperature, 0.6 K, the interferogram (a) shows several plane interfaces A, B, C (g) to be present. The plane A, of type $\langle 11\bar{2}0 \rangle$, shrank and disappeared by 0.8 K (b). Shortly afterwards the crystallographically equivalent plane B dramatically disappeared; compare (b) and (c), at 0.9 K. As the temperature continued to rise the facet C started to shrink and the crystal was observed to undergo a sudden change in external dimensions at 1.8 K. This transition reversed as the temperature was reduced again, and occurred a second time at the same temperature as the heating was recommenced. Apart from the changes in extremal dimensions of the crystal [Figs. 1(d)–1(f)], the transition is less obvious at first glance, because the facet C is almost parallel to the line of sight. To make the effect clearer, we show in Fig. 2 the area of the face C as a function of T, and the transition at 1.08 K is obvious. These observations suggest that the first two transitions in Table I occur at 1.08 ± 0.02 and 0.85 ± 0.05 K.

We found that ^{3}He fractions of the order of 10^{-4} or larger enhanced the flat faces, and eliminated curves in the interface.[6] This presents two problems: (a) How does ^{3}He stabilize facets? (b) How can such small concentrations of impurities affect the interface?

The answer to (a) is that ^{3}He pins the interface. This happens because ^{3}He is preferentially expelled from the solid phase by its zero-point energy. A capillary-wave description of the rough surface, following Ref. 4, shows that the time scale for interface fluctuations on an atomic scale is considerably shorter than that for diffusive motion of the ^{3}He impurities in the superflu-
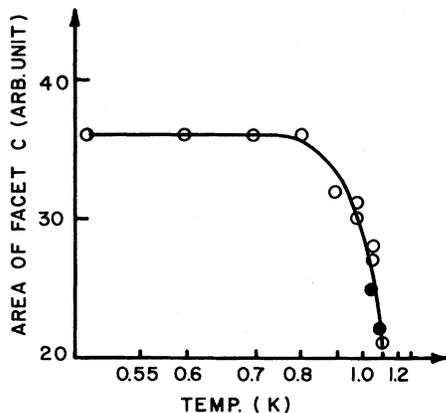
id. To understand (b), note[13] that the mean-square amplitude fluctuation of the rough interface $\langle Z(0)Z(r) \rangle$ is of the order $(T/2\pi\sigma_0\Omega)\ln(r/a)$. The interface flattens when the mean distance between the ^{3}He impurities, r, leads to fluctuations small compared to unity. The consequent logarithmic dependence on the concentration explains (b).

In conclusion, the concept of a roughening transition accounts for all the phenomena we have observed in ^{4}He hcp-superfluid interfaces.[27]

FIG. 2. The area of plane C as a function of T: open circles, increasing T; closed circles, decreasing T.

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Theory of the Surface Tension of Simple Liquids: Application to Liquid Metals

K. K. Mon and D. Stroud

Department of Physics, The Ohio State University, Columbus, Ohio 43210

This Letter presents an approximate closed-form solution to the density-functional theory for the surface tension and surface width of simple liquids. The results for both are in excellent agreement with experiments on Ar over wide range of temperature and density. The theory is extended via an ion density-functional formalism to simple metals. Good agreement is obtained for the surface tensions of ten liquid metals and for the surface width of liquid Hg.

The thermodynamics and structure of liquid-vapor interfaces are subjects of intensive experimental and theoretical interest.[1] Although various theoretical methods have obtained very good agreement with experiment for the surface tension of liquid Ar,[1,2] it is commonly believed that the surface free energies of liquid metals are dominated by electronic effects and that, therefore, theoretical techniques which are successful in treating simple nonmetallic surfaces cannot be used for metals. The work of Evans and Kumaravadivel,[3] which extends the theory of Lang and Kohn[4] to liquid metals, does lead to agreement with the surface tension in several cases but the predicted widths for the transition profile are small compared with those of most other fluids.

In this Letter we present a simple and quite successful theory for the surface tension τ and width l of simple liquids, both metallic and nonmetallic. It involves a number of approximations which allow one to solve a density-functional theory in closed form and obtain simple expressions for τ and l. The results for both are in excellent agreement with experiment for liquid Ar over wide range of temperature and reproduce the scaling results of Fisk and Widom.[5] The results for the surface tensions of liquid metals are in good agreement with experiment, for all metals studied. The surface widths emerging from the calculations are of the order of one to two atomic layers in all cases and for Hg, the one
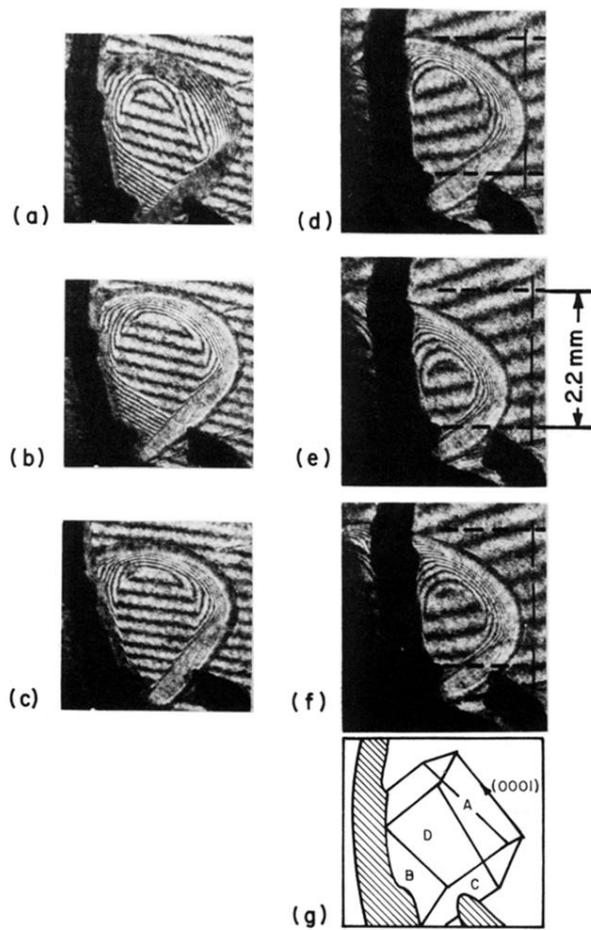
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FIG. 1. Equilibrium holographic interferograms of the ^{4}He crystal at constant volume: (a) at 0.6 K; (b) at 0.8 K below the lower transition; (c) at 0.9 K above the lower transition; (d) at 1.06 K before the upper transition; (e) at 1.09 K after the upper transition; (f) at 1.06 K after the reversal of (d) → (e); (g) schematic of the planes in (a) (Ref. 28).