# The interface in the problem of equilibrium crystallization

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The effect of quantum and thermal fluctuations on ordering of vicinal faces on the liquid-solid helium interface is examined. The consequences of this ordering, which are of experimental interest, are discussed.

Although the problem of equilibrium crystallization was stated by Wulff[1] (see also Ref. 2) at the beginning of this century, the only experimental study for which this theory can be used has been identified only recently. This is the interface between liquid and solid helium, experimental studies[3–5] of which were initiated by Andreev and Parshin.[6] Andreev and Parshin[6] made use of the dissipation-free nature of the melting at low temperatures and constructed a hydrodynamic theory of the motion of the surface. There remains, however, the problem of the microstructure of the surface and its state diagram. In this paper, we extend the theory of a crystalline face, formulated by Landau[7] for $T=0$ and for the simplest faces, to the case of a more complicated geometry and $T\neq 0$. An important problem discussed in recent years[8–10] is the following: can quantum fluctuations make the surface rough at $T=0$? We start from the model of the vicinal [with respect to (0,0,1)] face, constructed with steps of unit height. To simplify the problem we shall examine a cubic crystal, instead of a hexagonal one.

The quantum model of a single step is described by the spin ($S=1$) Hamiltonian

$$\mathcal{H}=\sum_m \{-\lambda(S_m^+ S_{m+1}^- + S_m^- S_{m+1}^+)+\Delta(S_m^z)^2 - hS_m^z + V(n) S_m^z S_{m+n}^z\}, \tag{1}$$

where $m$ enumerates the lattice sites along the step, the projections $S^z=\pm 1$ correspond to inflections in different directions, and the projection $S^z=0$ corresponds to the absence of an inflection on a step. The quantity $\lambda$ is equal to the amplitude of "attachment" of a particle to a step or drift away from it. The quantity $\Delta$ is the energy of a single inflection. The field $h$ controls the orientation of the step, while $V(h)$ is the energy of interaction of the inflections. Figure 1 shows the state diagram of the model, described by Hamiltonian (1). In the shaded region, the step is ordered, i.e., it is pinned by a definitive valley of the potential relief. In the case of high quantization $\lambda/\Delta \gg 1$, the step is smeared due to diffusion, and its fluctuation displacement by a distance $L$ is

$$\langle u^2\rangle \approx \frac{a}{4\pi}\frac{\hbar}{Ms}\ln\left(\frac{L}{a}\right). \tag{2}$$

Equation (2) can also be interpreted as the average displacement of a one-dimensional string with a predominant orientation under the action of zero point oscillations. This makes it possible to describe the factors in front of the logarithm: $M$ is the mass of the helium atom, and $s$ is the velocity of "sound" along the string.

  

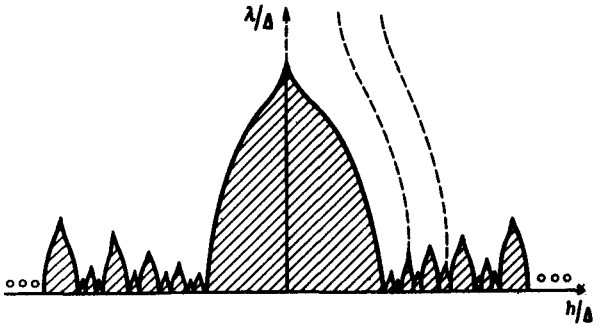


FIG. 1. State diagram described by Hamiltonian (1). The dashed curves indicate the lines of constant slope of the steps.

In a system of steps with average distance $l$ between them, the quantity $L$ in (2) must be replaced by $l$. These zero-point oscillations are not large. An estimate of the factor in front of the logarithm in (2) gives the value

$$\sim 0,1 a^2 T_\lambda / \theta_D ,$$

where $T_\lambda$ is the temperature of the $\lambda$ transition, and $\theta_D$ is the Debye temperature. Our simple estimates confirm the results obtained in Refs. 8–10 concerning the long-range order in the system examined at $T = 0$. For small oscillations of the steps, the periodic potential $V_0$ for the steps plays the role of the volume of the crystal. At $T = 0$, this system can be fixed by a periodic potential. However, a transition can occur to the unattached state (depinning). This is due to the classical interaction of the steps. The role of quantum fluctuations reduces to renormalization of the periodic relief. Making the assumption that the distribution of displacements with zero-point oscillations are Gaussian, we obtain the following expression for the renormalization of the potential:

$$\frac{V_R}{V_0} \sim \exp\left(-\frac{2\pi^2}{a^2} < u^2 >\right) = (a/l)^{(\pi/2)(T_\lambda/\theta_D)} .$$

The interaction decreases in a power-law fashion (see Refs. 7 and 11)

$$U(l) \sim (a/l)^{\Delta} \qquad (\Delta \geqslant 2).$$

For the stability commensurate with the volume of the structure, it is necessary that

$$V_R \geqslant U''(l).$$

For $T_\lambda/\theta_D < 1$ this is obviously satisfied for large values of $l$. Thus we have shown that vicinal faces are also stable near the face that is stable at $T = 0$. Our arguments are valid for vicinal faces of the form $(k,m,n)$ and $k \ll m \ll n$.

At $T = 0$, the problem of the optimal stepped arrangement of the interacting steps on the vicinal face $(0,m,n)$ $(m \ll n)$ is equivalent to the problem of the structure of a $1D$ interacting lattice gas with a fixed particle density $c = m/n$. The latter problem was solved in Refs. 12 and 13. Another formulation of the problem of the lattice gas with fixed chemical potential $\mu$ was used in Refs. 14 and 15. We are interested precisely in

this formulation of the problem, because the distance from the wall of the vessel, in the case of the interface, plays a role analogous to the chemical potential. We shall present the results of the solution of this problem.

On the $\mu$ axis, each rational $c$ corresponds to the finite segment

$$\Delta\mu(c) \sim d^2U/dp^2 \mid_{p=1/c}.$$

These values of $\Delta\mu$ are proportional to the dimensions of the flat sections of the face with a period $p$. The whole ensemble of these segments completely fills the entire $\mu$ axis.[15] The hierarchy of periodic phases which arises in the face can be described as a branching sequence. It begins with the main sequence, $\langle n\rangle \equiv (0,1,n)$. Each bifurcation occurs according to the following rule: a dimmer phase $\langle AB\rangle$ appears between neighboring phases $\langle A\rangle$ and $\langle B\rangle$ of the branching sequence and in the next bifurcation, the dimmer phase is the neighboring phase for the phases $\langle A\rangle$ and $\langle B\rangle$.

Because the interface is finite, only some faces can exist even at $T=0$. The size of the section proportional to the $\Delta\mu$ must be greater than the period of the structure. An estimate shows that the maximum possible periodicity does not exceed 20–30 lattice constants.

For any finite temperature, only a finite number of jumps remains in the derivative of the surface energy $dE/dc = \mu(c)$. This means that smooth and rough sections alternate on the face. It is convenient to solve the problem at $T \neq 0$ by the transfer-matrix method, analogously to the way this was done in Ref. 16. The transfer-matrix method reduces the problem of the statistics of a system of steps to the study of the ground state of a $1D$ quantum Hamiltonian. The role of the time axis here is played by the average direction of the steps, while the steps themselves are treated as world lines of fermions. The state diagram in the variables $\mu, Z = \exp(-E_0/T)$ is shown in Fig. 2. It goes over in the limit $T \to 0$ to the bifurcation sequence described above. An incommensurate phase exists between two neighboring phases $\langle A\rangle$ and $\langle B\rangle$ with $Z < \min(Z^c_{\langle A\rangle}, Z^c_{\langle B\rangle})$ ($Z^c_{\langle A\rangle}$ is the critical value of $Z$ of the ordered structure $\langle A\rangle$). For a fixed "time," the spatial arrangement of the steps looks like a disordered sequence of periods $A$ and $B$ with fixed concentrations of the periods. As the temperature is decreased, ordering appears in this sequence: a dimmer phase $\langle AB\rangle$ with period $l_{AB} = l_A + l_B$ is formed. The critical, behavior of the dimmer phases is universal. This behavior is described by the equations

$$T^c_{<AB>} Z^c_{<AB>} = \frac{1}{2}\frac{d^2U}{dp^2}\mid_{p=l_{AB}}, \tag{3}$$

$$\Delta\mu_{<AB>} = 4\pi l_{AB}\frac{d^2U}{dp^2}\mid_{p=l_{AB}} \exp\left(-\frac{\pi^2}{2\sqrt{2}\sqrt{(Z^c_{<AB>}-Z)/Z^c_{<AB>}}}\right). \tag{4}$$

The critical behavior of the main sequence $\langle n\rangle \equiv (0,1,n)$ does not differ qualitatively from that described above.

We shall also write an equation for the surface energy, which is valid in the temperature range $Z^c_{\langle n,n+1\rangle} \lesssim Z \lesssim Z^c_{\langle n\rangle}$:

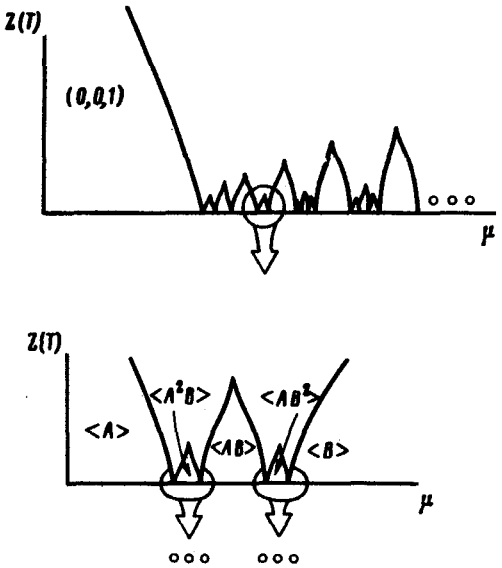


FIG. 2. State diagram of a statistical 2D system of steps. The fine structure of the state diagram is shown at bottom.

$$E(c = \mathrm{tg}\theta) = \mathrm{tg}\,\theta\left((n+1)\,U(n) - n\,U(n+1) + \epsilon_1 - 2TZ\,\frac{\sin\pi\{\mathrm{ctg}\,\theta\}}{\pi}\right), \qquad (5)$$

where $\{X\}$ is the fractional part of $X$, and $\epsilon_1$ is the step-formation energy (5). The jump $\Delta\mu$ which determines the spatial dimensions of the phase $\langle n\rangle$ and which is determined from (5) is

$$\Delta\mu(c = 1/n,\, T) = \Delta\mu(c = 1/n,\, T = 0) - 4nTZ(T). \qquad (6)$$

The transition temperatures $T^c_{\langle A\rangle} \sim (\ln l_A)^{-1}$ [see Eq. (3)] for a power-law interaction. So far, only the principal faces on the interface in helium have been found. The experimental observation of a 60-degree face by Wolf *et al.*,[17] which has an ordering temperature $T_0 \sim 0.3$ K, makes it possible to estimate the ordering temperature of the vicinal faces near (0,0,1):

$$T \sim T_0/\ln l \lesssim 0.1\,\mathrm{K}.$$

It is difficult to observe sharp features of the type (4) in experiments[1] using optics, because the limit of optical resolution $\sim 1$ $\mu$m is already reached at $(T_c - T)/T_c \sim 1/50$.

Most of our results refer to the case of cylindrical geometry in the field of gravity. The axis of the cylinder is assumed to be horizontal. The problem of determining the shape of the interface can be solved analytically (see also Ref. 18). The shape of the equilibrium surface depends on the dimensionless parameters, which are the edge angles and the quantity

$$G = \gamma^{-1} g\,\Delta\rho\,L^2,$$

where $g$ is the acceleration of gravity, $\Delta\rho$ is the difference of the densities of liquid and solid helium, $\gamma$ is the surface tension on the interface, and $L$ is the linear size of the interface. By arranging the axis of the nucleus in an appropriate manner, it is possible to increase the area of the fixed face by a factor of $\sqrt{G}$.

The edge angle is a discontinuous function of the angle between the axis of the crystal and the surface of the wall. The number of discontinuities and their magnitude decrease with increasing temperature. This effect could have been observed by rotating the cylinder around its axis.

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The results of the calculations will be published elsewhere.

[1]G. Wulff, Z. Kristallogr. und Mineralogie **34**, 449 (1901).
[2]C. Herring, Phys. Rev. **82**, 87 (1951).
[3]K. O. Keshishev, A. Ya. Parshin, and A. V. Babkin, Pis'ma Zh. Eksp. Teor. Fiz. **30**, 63 (1979) [JETP Lett. **30**, 56 (1979)].
[4]J. E. Avron, L. S. Balfour, C. G. Kuper, J. Landau, S. G. Lipson, and L. S. Shulman, Phys. Rev. Lett. **45**, 814 (1980).
[5]S. Balibar and B. Castaing, J. Phys. (Paris) Lett. **41**, L 329 (1980).
[6]A. F. Andreev and A. Ya. Parshin, Zh. Eksp. Teor. Fiz. **75**, 1511 (1978) [Sov. Phys. JETP **48**, 763 (1978)].
[7]L. D. Landau, in: Symposium in Honor of 70th Birthday of Academician A. F. Ioffe, Izd. Akad. Nauk SSSR, 1950, p. 44.
[8]D. S. Fisher and J. D. Weeks, Phys. Rev. Let. **50**, 1077 (1983).
[9]E. Fradkin, Phys. Rev. B **28**, 5338 (1983).
[10]S. V. Iordanskiĭ and S. E. Korshunov, Pisma Zh. Eksp. Teor. Fiz. (in press).
[11]V. I. Marchenko, Pis'ma Zh. Eksp. Teor. Fiz. **35**, 459 (1982) [JETP Lett. **35**, 567 (1982)].
[12]J. Hubbard, Phys. Rev. B **28**, 5338 (1983).
[13]V. L. Pokrovsky and G. V. Uĭmin, J. Phys. C **11**, 3535 (1978).
[14]P. Bak and R. Bruinsma, Phys. Rev. Lett. **49**, 249 (1982).
[15]S. E. Burkov and Ya. G. Sinaĭ, Usp. Matem. Nauk **38**, 205 (1983).
[16]G. V. Uĭmin and V. L. Pokrovsky, J. Phys. (Paris) Lett. **44**, L865 (1983).
[17]P. E. Wolf, S. Balibar, and F. Gallet, Phys. Rev. Lett. **51**, 1366 (1983).
[18]J. E. Avron, L. S. Balfour, and R. Zia, J. Stat. Phys. (in press).

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