CRYSTALLIZATION WAVES IN HELIUM

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Both theoretical and experimental evidence for a new quantum state of the ^{4}He liquid–solid interface are presented. The existence of this state implies the possibility of exactly non-dissipative crystallization and melting. Weakly damped oscillations of the interface due to periodic melting and crystallization – crystallization waves – may propagate along the interface under these conditions. The spectrum of these waves is measured and the frequency and temperature dependences of their attenuation are determined. At $T \leq 1$ K the interfaces of any crystallographic orientations are believed to be in a quantum state, except the two faces identifiable with respect to symmetry.

1. Introduction

As is well known (see refs. [1–3]) a macroscopically homogeneous surface of any classical crystal in equilibrium with liquid or vapour can be atomically smooth or atomically rough. In the rough state there are many thermodynamically equal surface defects, such as steps, kinks, adatoms, etc. The smooth surface might be considered to be a two-dimensional crystal with few defects, and the rough surface a two-dimensional liquid. Naturally such a liquid might exist as a thermodynamically equal substance at sufficiently high temperatures only. As the temperature drops such a liquid should, in the long run, "freeze" (the so-called roughening transition), just as normal three-dimensional liquids do. In other words, the surface of any classical crystal at absolute zero should be an atomically smooth one without any defects. This statement is inspired by the fact that any classically rough surface does not exist at absolute zero in equilibrium because its entropy is not zero.

The situation can be changed by quantum effects [4]. Namely, because of the large zero-point motion of helium atoms an atomically rough surface can remain in the liquid state down to absolute zero – just as liquid helium does at low pressures. Naturally, this state is not a rough one in the classical sense, but its quantum analog. At absolute zero all kinds of roughness would be delocalized and collectivized, their motion would be exactly coherent just like the motion of particles in a normal superfluid liquid. This coherence implies in particular that growth and melting of the crystal with such a surface at absolute zero would be exactly nondissipative, without any disturbance of phase equilibrium (of course the absence of energy dissipation in volumes of the two phases in equilibrium is also required). Because of the ease in obtaining phase equilibrium such a situation opens up great possibilities for the investigation of capillary phenomena in crystals. Firstly, the direct measurement of the surface energy of crystals by investigation of their equilibrium shape becomes possible. A new phenomenon then appears to be possible under these conditions: weakly damped oscillations of a surface due to periodic melting and crystallization while the crystal itself stays undeformed and immoveable. These oscillations, termed crystallization waves, might be of any amplitude, restricted only by their crystal size. Recently, this phenomenon was observed experimentally [5]. Below, the conditions required for the new quantum rough state, its microscopic structure and macroscopic properties are considered and then the results of experimental investigation of crystallization waves [6] are presented.

2. Theory

Delocalization of particles in a quantum crystal means that the microscopic structure of such a phase boundary cannot be represented in the literal way, as is done for classical crystals. However, such concepts as a surface step and a kink in the step can also be introduced in the quantum case. It is sufficient to use the general properties of a boundary which follow from the symmetry of phases in contact. For example, the surface step corresponds to such a state of the boundary in which its positions at infinity (to the right and left in fig. 1) are shifted by an elementary translation vector of the crystal and the energy of the system has the lowest possible value. In view of the periodicity of the crystal and homogeneity of the liquid, a shift by the translation vector transforms the boundary to an equivalent position and, therefore, a step is a linear defect on the surface. It is this property of the step that is important in our analysis. It should be noted that the concepts of a surface step (and, similarly, a kink in the step) can be introduced in this way for the surface of any integer Miller indices.

The state of a step is governed by the configuration of the kinks, and the kinks have two opposite "signs" (fig. 1). Each kink can be regarded as a point defect on a step. Let us consider a step on the surface in equilibrium at $T = 0$. The displacement of the kink by a translation vector along the step involves the transfer of a particle (helium atom) from one phase to another. Such transitions occur with a finite probability, even at $T = 0$, provided by processes similar to quantum tunnelling. The kink energy
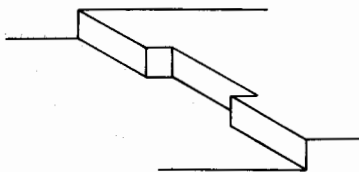

Fig. 1. An elementary step with two kinks.

does not change as a result of this displacement because the chemical potentials of the phases in equilibrium are equal. Therefore, like other point defects in quantum crystals [7] such a kink behaves like a delocalized quasiparticle. Its state is defined by the quasimomentum p. The energy of a localized kink, ε_0, is expected to lie approximately in the middle of the energy band $\varepsilon(p)$. Let p_0 be the value of the quasimomentum corresponding to the bottom of the energy band. At $T = 0$ this state is stationary (and ground) for an isolated kink and the velocity of the kink is zero. Stationary states of close energies $(p \to p_0)$ correspond to finite kink velocities. In other words, the transfer of matter from one phase to the other in this case is a coherent process occurring without any energy dissipation. It is important to note also that the states under consideration are not separated from the ground state by an energy gap. Thus, an isolated kink is an example of a system having the stationary states whose energies can be arbitrarily close to the ground state, characterized by a continuous flux of matter from one phase to the other.

There are two possibilities for further consideration. If the energy corresponding to the bottom of the energy band $\varepsilon(p_0)$ is positive, no kinks occur in the step's ground state, i.e. the step is atomically smooth at $T = 0$. But if the width of the energy band Δ is sufficiently great (roughly speaking, if $\Delta > 2\varepsilon_0$), then $\varepsilon(p_0) < 0$, i.e. the total energy of the step decreases with the appearance of kinks with $p = p_0$. If it is the case for kinks of either sign, the atomically smooth state becomes unstable with respect to the creation of pairs of kinks of opposite signs (the creation of kinks of only one sign is impossible at fixed step orientation). Since the interaction between kinks is important, generally speaking, only over atomic distances, an increase in kink concentration in a step thus reduces the energy of the latter until the minimum is reached at the concentration of the atomic order. The essential property of such a step is the existence of stationary states which are close in energy to the

ground state and involve continuous step motion accompanied by the transfer of particles between the phases. The nature of these stationary states becomes clear from consideration of possible types of kink collision. In the case of two kinks of the same sign the only possible elastic process is the exchange of their quasimomenta (fig. 2a); however, if the signs of the kinks are opposite, besides the latter there is also the possibility of a "jump" to the next row wherein each kink conserves its quasimomentum (fig. 2b). It is the second type of processes that give rise to the stationary states under consideration. Such a "quantum rough" step thus appears to be completely delocalized at the crystal surface.

The total energy of isolated quantum rough step β (per unit length equal to the elementary translation vector along the step) can be either positive or negative depending on whether the positive energy of the "bare" atomically smooth step β_0 is sufficient to compensate for the negative energy of delocalized kinks including the energy of its interaction. If $\beta > 0$ for arbitrary step orientation on the given surface, its ground state does not involve any steps and at $T = 0$ the surface remains in the classical atomically smooth state. If $\beta < 0$ for any pair of steps having the opposite orientations (of course more complex configurations are also possible), an atomically smooth surface appears to be unstable with respect to such step creation. As a result, an equilibrium surface should become a type of two-dimensional liquid consisting of delocalized

Fig. 2. Possible types of kink collisions: (a) the "normal" collision; (b) the collision with a "jump" in the next row.

steps of various configurations, including closed steps of finite length. It is important to note that the number of steps of each kind in such a liquid is not fixed and is defined by the condition of minimal energy. Therefore the energy of a step (including the energy of its interaction with the others), which is the derivative of the total energy with respect to the number of steps, vanishes in equilibrium. Such a state of the surface might be called "quantum rough".

This vanishing of the step energy β is connected closely with equilibrium crystal faceting. Indeed, according to Landau [8] if β is finite then the derivative $\partial\alpha/\partial\varphi$ of the surface energy α with respect to the crystallographical angle φ exhibits a finite jump proportional to β when φ corresponds to the initial surface orientation. This discontinuity of the function $\partial\alpha/\partial\varphi$ necessarily gives rise to flat parts in the equilibrium shape of a crystal and these parts increase in size with the jump of the derivative $\partial\alpha/\partial\varphi$. At $T = 0$ in the classical case such a property is intrinsic for a surface of any integer Miller indices. Thus, any classical crystal is always completely faceted in equilibrium at $T = 0$. This statement is in natural agreement with the above conclusion that the surface of a classical crystal has to be atomically smooth under these conditions.

As far as crystals with quantum rough surfaces are concerned, according to what was said above their surface energy is a smooth function of crystallographic angles. Therefore, crystals with quantum rough surfaces of any orientations are not faceted, even at $T = 0$. It is clear also that the situation is possible when the surfaces of some singular orientations (corresponding to the mostly close-packed faces, because these are expected to have maximum values of β_0) stay in the atomically smooth state while the rest are quantum rough. In this case the equilibrium shape of a crystal involves a finite number of flat parts ringed by a rounded surface. Such a situation, as we shall see below, takes place for the hcp-^{4}He.

Thus, an atomically rough state can occur

either (in classical crystals) because of thermal reasons, i.e. at sufficiently high temperatures, or (in quantum crystals) because of the effect of quantum delocalization. One might say that the stronger the quantum effects the lower the number of crystal faces that stay in the classical atomically smooth state at $T = 0$, i.e. a lower number of flat parts arise in equilibrium faceting.

Naturally, the microscopic dynamics of crystallization (and melting) are believed to be completely different processes for the two different surface types. The growth of a crystal with a quantum rough surface occurs both by an increase in the surface bounded by each step and by the formation of new atomic layers on collision of two steps. The latter process is similar to a jump into the next row with the collision of two kinks). It is important to note that these and reverse processes give rise to stationary states of the system which are arbitrarily close in energy to the ground state and the continuous growth or melting of a crystal occurs via these states. The motion of a phase boundary at absolute zero thus occurs without disturbing the phase equilibrium. In other words, the kinetic growth coefficient K (defined by the formula $V = K\Delta\mu$, where V is the velocity of the boundary and $\Delta\mu$ is the difference between the chemical potentials of the phases in contact) becomes infinite at $T = 0$ for a crystal with this type of surface. On the other hand, an atomically smooth surface's growth coefficient at $T = 0$ appears to be equal to zero, even taking into account the possibility of quantum under-barrier tunnelling [9].

At finite temperatures the motion of the quantum rough boundary is accompanied by dissipation due to its interaction with a volume's thermal excitations. Let us consider the dissipation due to phonons. The phonons incident to the boundary from the liquid give rise to the pressure $P_{ph} \sim E_{ph} \sim nT^4\theta^{-3}$ (n is the number of atoms per unit volume and θ is the Debye temperature). In equilibrium, i.e. when the boundary is motionless, this pressure is compensated by the pressure of the crystal. If the boundary

moves with some velocity V, then the difference of pressures opposite to the motion arises due to the Doppler effect, $\Delta P_{ph} \sim E_{ph} V c^{-1}$ (c is the velocity of sound). As a result, the energy dissipated per unit time and per unit surface area is of the order of $V^2 c^{-1} E_{ph}$. On the other hand, the same dissipated energy is equal to $\dot{N}\Delta\mu$, where $\dot{N} \sim nV$ is the number of atoms transferred per unit time from one phase to the other. Hence, we find that

$$K \sim c\theta^3 T^{-4}. \tag{1}$$

We can thus see that the growth coefficient becomes infinite in the limit $T \to 0$ which it approaches proportionally to T^{-4}.

Phonons are known to be the main type of thermal excitations in superfluid helium at temperatures below approximately 0.5 K. At higher temperatures rotons are believed to provide the major part of the total energy dissipation. In this roton region we now have the exponential dependence $K \propto \exp(\Delta_r/T)$, where Δ_r is the roton gap.

As for the ^{3}He case, that at the temperatures when liquid ^{3}He behaves as a Fermi liquid, the main dissipation mechanism is the interaction of a moving boundary with Fermi excitations. The energy dissipation is of the order of $p_F \cdot nV \cdot V$, where p_F is the momentum on the Fermi surface, which corresponds to a temperature-independent growth coefficient $K \sim p_F^{-1}$. A similar estimation could be made for the case of dilute solutions of ^{3}He in ^{4}He (the concentration of ^{3}He in liquid phase $n_3/n \ll 1$). In this case the energy dissipation, being due to the interaction of the moving boundary with impurities, is of the order of $n_3 p_3 V^2$, and the growth coefficient $K \sim nn_3^{-1}p_3^{-1}$ (p_3 is the characteristic momentum of an impurity). Comparing with (1) we see that at low temperatures the growth coefficient of ^{4}He appears to be highly influenced by small ^{3}He fractions.

As we have seen above, the quantum rough state of a surface gives rise to the possibility of

exactly non-dissipative growth and melting at $T = 0$. If this is the case, then weakly damped oscillations, similar to normal capillary waves, may propagate along the surface at sufficiently low temperatures. Indeed, any deflection from the equilibrium crystal shape results in an increase in surface energy. Therefore any disequilibrium crystal shape should change by crystallization or melting reducing its surface energy. On the other hand, because the densities of the two phases are different, growth and melting of a crystal give rise to a motion of a liquid, i.e. an increase in the kinetic energy of the system. As a result oscillations of an interphase boundary should occur if the total energy dissipation is small. The spectrum of these oscillations termed, "crystallization waves" can be found easily in the long-wave limit where the compressibilities of the two phases are negligible. It is important to note that no models of the surface's microscopic structure are required for this. Taking into account gravity g and damping due to the finite growth coefficient K the spectrum of plane waves $\omega(k)$ is given by

$$\omega^2 - \frac{\tilde{\alpha}\rho_2}{(\rho_1 - \rho_2)^2} k^3 - \frac{\rho_2 g}{\rho_1 - \rho_2} k$$
$$+ \frac{\rho_1 \rho_2}{(\rho_1 - \rho_2)^2} \frac{i\omega k}{mK} = 0, \tag{2}$$

where $\tilde{\alpha} = \alpha + \partial^2\alpha/\partial\varphi^2$, φ is the angular variable in the plane of k, ρ_1 and ρ_2 the densities of the solid and liquid phases, respectively, and m the mass of an atom. As we can see, the propagation velocity of the oscillations under discussion is much less than the velocity of sound and the compressibilities of the two phases may indeed be neglected. The gravitational term in eq. (2), as for normal capillary waves, need be taken into account only for the lowest frequencies, when the wavelength becomes of the order of the capillary constant (~ 1 mm for ^{4}He).

Like the normal capillary waves, the oscillations described by eq. (2) are unstable with respect to decay of one quantum into two of

lower energy. Therefore, these oscillations are characterized by finite damping, even at $T = 0$. This damping, however, is very small for macroscopic wavelengths. At finite temperatures and for low-frequency oscillations the most important damping mechanism is due to the finite growth coefficient K. For the case of weak damping ($\kappa \ll k$, where κ is the inverse damping length) we find from eq. (2):

$$\kappa = \tfrac{1}{3}\rho_1\rho_2^{1/3}(\rho_1 - \rho_2)^{-2/3}\tilde{\alpha}^{-2/3}(mK)^{-1}\omega^{1/3} \tag{3}$$

(here we have ignored gravity).

In the case of ^{3}He, within the normal Fermi liquid range, the use of the above estimate of K leads to the conclusion that the oscillations in question are strongly damped at all frequencies. naturally, similar estimates for the case of a classical atomically rough surface lead to the same conclusion. As for the case of an atomically smooth surface, any linear oscillations similar to capillary-crystallization waves are believed to be completely impossible.

Let us note, finally, that the crystallization waves quanta are elementary excitations of the ^{4}He quantum rough surface at low temperatures. They are responsible for the temperature dependence of the surface energy. Since the frequency is proportional to $k^{3/2}$, the temperature-dependent component of surface energy is proportional to $T^{7/3}$, as in the case of capillary waves at the liquid–vapour interface. However, numerical estimates show that the current experimental accuracy is not sufficient to detect this temperature dependence.

3. Experiment

In our experiments the optical ^{3}He cryostat was used, which is a modification of Shal'nikov apparatus [10]. Four pairs of plane-parallel windows enabled the observation of crystal growth, to photograph and to accomplish simple optical measurements. The ^{4}He crystals were grown in a

metallic cell $12 \times 15 \times 28$ mm (1, fig. 3), having two plate glass walls, which allowed the crystallization process to be observed throughout the cell. A capacitor designed to excite surface oscillations was mounted on one of the metallic walls inside the cell (2), consisting of two copper wires in a capron insulator $\phi = 30 \mu$m wound on a textolite plate. The maximum electrical field intensity was $2–3 \times 10^6$ V/cm. The filling capillary (3), electrical leads (4) and copper heat conductor (5) to the ^{3}He bath are also shown in fig. 3. The capillary also had good thermal contact with the ^{3}He bath. The cell temperature was measured with a carbon resistor (6), which was mounted outside the cell in thermal contact with Cd superconducting reference point (7). With the cell full the total heat leak to the ^{3}He bath was less than 10^{-4} W including the heat leak by radiation through the windows.

The helium crystals were grown at constant temperatures (0.4–1.3 K), pressures (25 atm) and controlled helium flow into the cell. Visual observations for the ^{4}He growth processes discovered a great variety of crystal shapes and the growth dynamics depending on the temperature and nuclei positions and orientations. It is important to note that the growth dynamics and the crystal shape are highly influenced by gravity. As a rule, at temperatures lower than 1.2 K, crystallization began at a single random point and during growth the crystal was always faceted more or less: the faster the growth rate and the

Fig. 3. The experimental cell.

lower the temperature, the sharper the facets. Observed faceting always corresponds to hexagonal prisms represented by a greater or lesser number of its facets depending on the nucleus position and orientation and the crystal size at the moment of observation (fig. 4). Bearing in mind the symmetry of ^{4}He crystals (hcp-structure) we may identify the basal facets of the prism as (0001) crystal planes and the lateral facets as the $\{11\bar{2}0\}$ type (or, alternatively, as the $\{10\bar{1}0\}$ type). Fig. 4 very clearly shows all the characteristic features of the ^{4}He crystal's low-temperatures growth: (a) the complete faceting during rapid growth ($\geqslant 0.1$ mm/sec), (b) the appearance of rounded regions in the upper part of the crystal surface during slow growth, and (c) the forming of the characteristic meniscus. On the contrary, during a sufficiently rapid melting the crystal's shape becomes drop-like.

The above features of the growth of ^{4}He crystals have also been observed by Landau and his co-workers [11] using optical-holographic techniques. All these features may be understood by the anisotropic growth coefficient. Precisely, this coefficient reaches its minimum values at the directions normal to the faces of the growth prism and has cusps in these directions [12]. Moreover, direct visual observation of the ^{4}He growth processes enable us to draw some conclusions about the temperature dependence of the growth coefficient. At the growth prism faces, especially at the (0001) face, this coefficient drops rapidly with decreasing temperature. On the contrary, the other faces exhibit a rapid growth coefficient increase with falling temperature. The latter fact is believed to prove that the quantum rough state is being realized at every face, except the singular faces (0001) and $\{11\bar{2}0\}$ (or $\{10\bar{1}0\}$). These singular faces are believed to be atomically smooth at low temperatures.

The observations of the equilibrium crystal's shape have fully confirmed this conclusion. In general, the equilibrium surface of a large crystal exhibits a convex meniscus. The measurement of
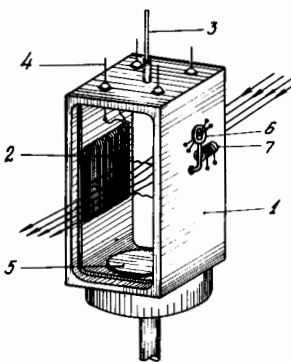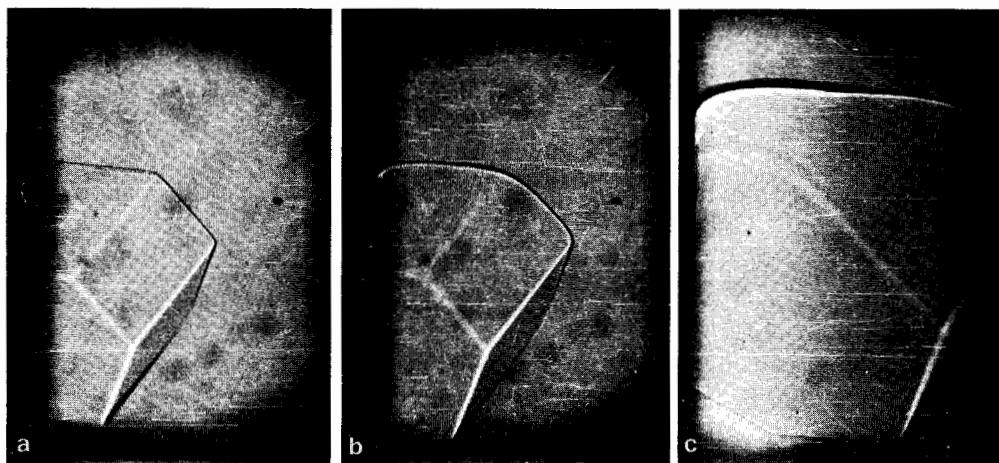
Fig. 4. Different stages in the growth of one of the crystals at 0.5 K.

the meniscus parameters enables us to estimate the surface energy of the interphase boundary α, which appeared to be 0.1–0.2 erg/cm^2, depending on the interface orientation. No temperature dependence was found up to the hcp–bcc transition (see also ref. 11).

When one of the singular faces is sufficiently close to horizontal the equilibrium meniscus includes a large flat part (fig. 5) the shape and size of which are very sensitive to its small inclines ($\sim 1°$). This plane is identical to corresponding singular face that becomes obvious after the regrowth of such a crystal. Of course, flat parts can be observed also in the surfaces of small crystals having arbitrary orientation (see also refs. 11 and 13). However, in such cases it is much more difficult to prove its flatness and equilibrium nature. According to our obser-
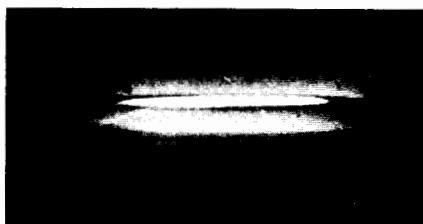
vations (see also ref. 13) the sizes of the flat parts decrease with increasing temperature. The temperature of its vanishing (roughening transition) seems to be 0.9 K for the lateral facets and 1.17 K for the basal facets. These data, however, cannot be considered sufficiently reliable because of the difficulty of obtaining a true equilibrium.

The relaxation of the large crystal shape to the equilibrium meniscus (nonsingular!) becomes more rapid if the temperature drops and appears to be oscillating at temperatures lower by approximately 0.7 K. The simplest means of exciting these surface oscillations is the mechanical vibration of the cell – just as in case of normal capillary waves at a liquid surface. Crystallization waves were found experimentally in this very way. As it turned out, even a small vibration of the cryostat was enough to excite visible oscillations of the surface. By tapping on the outer wall of the cryostat, the amplitude of the oscillations reached 1–2 mm. Fig. 6 shows a motion picture sequence of the excitation and damping of oscillations. The first frame shows the calm boundary between the solid (located below) and liquid phases. The subsequent frames demonstrate the behaviour of the boundary after a sharp blow on the outer wall of the cryostat. This

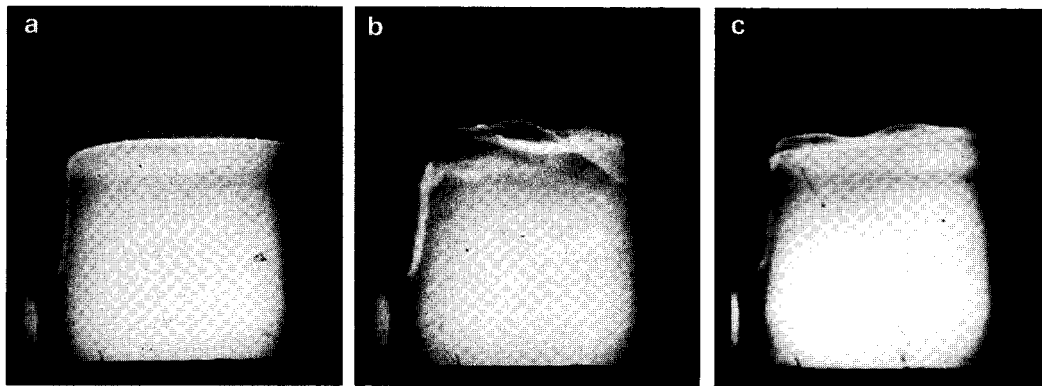Fig. 5. The meniscus with the (0001) facet nearly horizontal.

Fig. 6. Motion picture frames of the excitation and decay of crystallization waves at 0.5 K.

manner of exciting surface oscillations becomes ineffective at temperatures higher than approximately 0.7 K.

If the equilibrium meniscus includes one of the singular faces, surface oscillations can be observed at rounded parts only, while the flat part remains completely motionless (of course the outlines of this flat part oscillate due to oscillations of the rounded parts). Crystallization waves can be observed not only in such quasiequilibrium situations when a crystal fills the lower part of the cell, but also in situations when, for example, a crystal is hanging on a lateral wall of the cell (fig. 7). In the latter case oscillations can be clearly observed in the rounded regions (and only in them). All these data once more demonstrate the difference between a quantum rough and a classical atomically smooth surface's behaviour at low temperatures.

To measure the spectrum of the crystallization waves a capacitor mounted on one of the metallic walls inside the cell has been used (2 in fig. 3). A d.c. voltage at the capacitor gives rise to a visible contact angle change and corresponding ascent of the meniscus region close to the capacitor. An a.c. voltage of an appropriate frequency ω in addition to the d.c. voltage causes the crystallization wave excitation with the frequency ω and the wave vector k normal to the line of the crystal–capacitor contact. The detec-

tion technique includes a narrow laser beam, slowly scanning along the surface in the direction of k. The lower part of the beam is refracted by the slightly convex surface and deflects down significantly while the rest of the light falls on a photodetector. Thus, the detector's signal appears to be modulated periodically by the propagating surface wave. The amplitude of this modulated signal is proportional to the wave amplitude, whereas the signal phase is equal to the phase of the wave at a given point. This
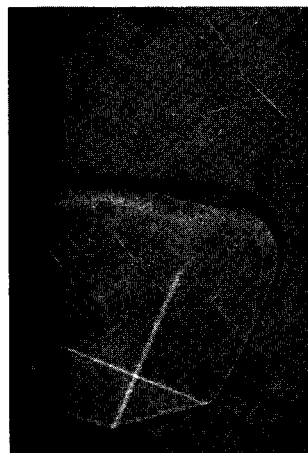

Fig. 7. A small crystal under quasiequilibrium conditions. The lower part of the crystal surface is completely faceted and immobile, whereas the upper part clearly oscillates due to the cryostat vibrations.

signal can be recorded (see fig. 8). The distance along the abscissa between zero points of the signal determines the wavelength, and the ratio of maximum neighbouring signals measures the damping.

Fig. 9 shows the measured spectrum of one of our samples at two different temperatures. The dotted line corresponds to $\omega \propto k^{3/2}$, the solid line is the theoretical dependence $\omega(k)$ according to eq. (2) with a gravitational term and $\tilde{\alpha} = 0.21\ \text{erg/cm}^2$ (the correction for damping is negligible). The data for other samples are the same excepting some shift of the curve corresponding to the change of $\tilde{\alpha}$ value. Thus, the $\tilde{\alpha}$ values from $0.097\ \text{erg/cm}^2$ (the dashed line in fig. 9) to $0.23\ \text{erg/cm}^2$ were fixed. These data show that the surface energy α is significantly anisotropic.

The measured frequency dependence of damping $\kappa(\omega)$ is presented in fig. 10. Clearly, the theoretical dependence $\kappa \propto \omega^{1/3}$ is well satisfied, i.e. the observed damping is really due to the finite growth coefficient K. According to the above discussion, at low temperatures $K \propto T^{-4}$, whereas at higher temperatures $K \propto \exp\,(\Delta_r/T)$. Fig. 11 shows the temperature dependence of $(mK)^{-1}$ calculated from the measured values of κ for three different samples.

We may conclude from Fig. 11 that at low temperatures damping drops with temperature too slowly compared with the phonon damping. However, it is necessary to take into account that
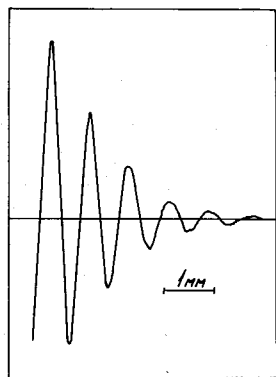
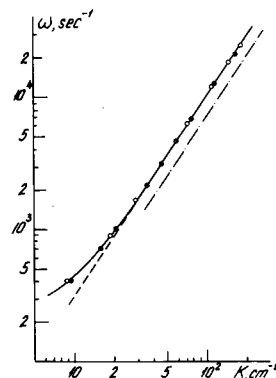

Fig. 9. Measured spectrum of crystallization waves: $\bigcirc$ $T = 0.360$ K; $\bullet$ $T = 0.505$ K.

in real experiments there are many reasons for a residual damping, i.e. finite damping even at arbitrarily low temperatures. These are different processes of the scattering of crystallization waves due to the crystal defects, e.g. nonlinear effects due to random vibrations of the apparatus, the influence of the real wave generator geometry and the real meniscus shape, etc. This residual damping could change significantly from one sample to another and could change with frequency in a way that depends on the major mechanism of scattering. From this point of view the discrepancy of the data for the sample N3 in fig. 11 at the two different frequencies can be understood.
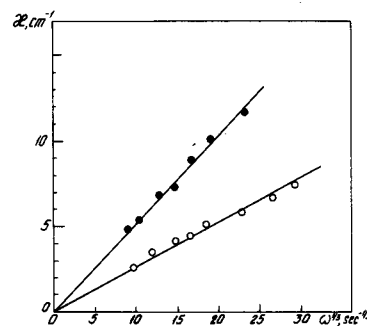
These considerations allow us to interpret the

Fig. 8. An example of the experimental record.

Fig. 10. The frequency dependence of the damping of crystallization waves: $\bigcirc$ $T = 0.360$ K; $\bullet$ $T = 0.505$ K. The sample is the same as that in fig. 9. The straight lines are $\kappa \propto \omega^{1/3}$.
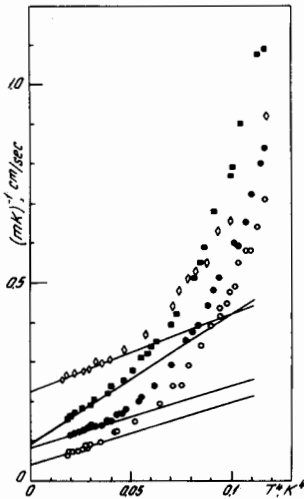
Fig. 11. The temperature dependence of the crystallization waves damping: sample No. 3, ○ 1118 Hz, ● 232 Hz, sample No. 4, ■ 827 Hz, sample No. 5, ◇ 837 Hz.

data of fig. 11 as follows. Let us consider the measured damping as the sum of three independent items: residual, phonon and roton:

$$\frac{1}{mK} = A(\omega) + BT^4 + C\,e^{-\Delta_r/T}. \qquad (4)$$

According to the above, the coefficient A could change from one sample to another and also could change with frequency. Because of strong anisotropy of the sound velocity in ^{4}He crystals phonon damping B can change
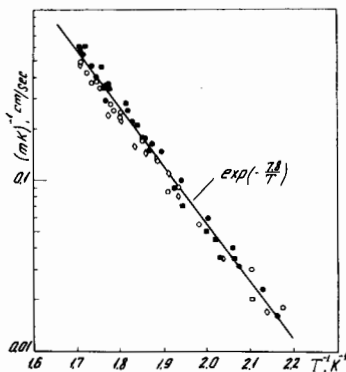

Fig. 12. The roton damping defined using eq. (4). The signs are the same that in fig. 11.

significantly depending on the crystal orientation. As far as the roton item C is concerned, it seems to be approximately the same for all the samples.

In accordance with this interpretation the straight lines are plotted in fig. 11 to show the sum of the first and the second items in eq. (4). Defined in such a way, the roton term is presented in fig. 12. It is clear that all the data lie on the same exponential, within experimental error. This fact seems to be the most serious argument supporting the above interpretation. The deduced value Δ_r (7.8 K) is slightly higher compared with the neutronographic data (7.0–7.2 K) [14]. However, this discrepancy could be imaginary since the coefficient C could depend on the temperature by some power law, similar to the variety of thermodynamical functions depending on roton spectrum (heat capacity, normal density, etc.).

Recently Castaing, Balibar and Laroche [15], investigating transmission of sound across the liquid–solid interface of ^{4}He, have measured a quantity equivalent to the growth coefficient and obtained an exponential dependence with Δ from 5 to 7.8 K for different samples. For the complete solution of the problem a theory is now needed, including the exact calculation of the roton damping and the damping due to the volume dissipative processes.

4. Conclusion

The existence of weakly damped oscillations of the ^{4}He crystal surface due to periodic melting and crystallization means that quantum delocalization is the determining factor for the helium crystallization processes at low temperatures. In essence we are here dealing with a new macroscopic quantum phenomenon – coherent phase transition. Indeed, the exact nondissipative crystallization is possible only in the presence of a special type of coherent motion in a two-phase system, opposite to the classical situation when the crystallization process is a

result of random transitions of individual particles from one phase to another. Of course, the current theory is not sufficient for the complete description of this phenomenon; in essence this theory covers its main point only.

Generally, with increasing temperature the significance of the quantum effects diminishes. It is interesting to estimate the temperature region where quantum effects are essential for the helium crystallization processes. For this purpose we use the following simple consideration. Quantum effects cannot be negligible if at a given temperature a wavelength region exists where the crystallization waves are weakly damped. The temperature T_0, when the damping becomes of the order of unity, even for wavelengths as short as the interatomic distance, can be estimated by the extrapolation of the data of fig. 12 and using eq. (3). Such an estimation gives $T_0 = 1.5$ K. In other words, up to the hcp–bcc transition (and perhaps higher) the processes occurring on the surface of helium crystals have essentially quantum features.

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