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Crystallization waves
In memory of Alexander Parshin

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Melting-freezing wave is a peculiar phenomenon taking place on a surface of quantum crystal. This wave can propagate due to the extremely fast growth dynamics of the suface and due to the non-dissipative mass transport provided by the superfluid surrounding a crystal. This short paper reviews the studies of crystallization waves starting from their theoretical prediction and the first experimental observation by Alexander Parshin and his collegues.

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In the year 2013 Alexander Parshin passed away in the age of 74. This sad event urges to recall the legacy of this outstanding scientist in physics who combined the elegancy of best experimentalists with deep theoretical understandings. Among various fields in physics where Parshin contributed there is one which is inseparably connected with his name, the physics of quantum crystal surface.

A crystal is called quantum crystal if the amplitude of zero-point motion of particles in its lattice is comparable with the lattice constant. The corresponding parameter is known as quantum parameter of de Boer [1], $\Lambda \sim (\hbar/a)(mU)^{-1/2}$, which is largest for light weekly interacting particles, like helium isotopes ^{3}He ($\Lambda \approx 0.5$) and ^{4}He ($\Lambda \approx 0.4$).

Due to the large zero-point motion of atoms, helium solidifies only under high pressure, ^{4}He at 25.3 bar and ^{3}He at 34.4 bar, being the only substance in nature whose liquid phase exists down to absolute zero. At low enough temperatures the latent heat of crystallization vanishes, and the liquid phase becomes superfluid and supports non-dissipative mass transport, which results in uniquely high mobility of solid-liquid interface of helium. Practically, helium crystal can grow/melt so fast that the wave of crystallization/melting sustained by the inertia of the liquid flow can propagate along the crystal surface.

The idea of the crystallization wave has appeared in 1978 in the famouse paper "Equilibrium shapes and oscillations of the surface of quantum crystals" by Andreev and Parshin [2]. In this paper the great importance of quantum fluctuations of the surface of helium crystals has been first recognized. Andreev and Parshin have argued in this paper that: (**i**) the short-range quantum fluctuations in the case of ^{3}He might be so strong that no facets are present on ^{3}He crystal surface at any temperature, and (**ii**) at low enough temperatures the mobility of the solid-liquid interface of helium becomes so high that the low-damping waves of crystallization/melting can propagate along the crystal surface.

The first statement was based on the idea of quantum delocalization of kinks on elementary steps on the facet. The kink thus behaves as a quasiparticle with the energy band $\Delta \approx 1$ K while the energy of the localized kink is much smaller, $\varepsilon_0 \approx 0.1$ K. As the activation energy of the isolated kink, $\varepsilon_0 - \Delta/2$, becomes negative, the elementary step becomes unstable with respect to the creation of kinks, and its free energy, in turn, becomes negative and the facet is blurred away by spontaneous creation of highly meandered steps, i. e. by the surface fluctuations. It has been shown later by Fisher and Weeks and by Parshin with collegues by more involved analisys that quantum fluctuations indeed can greatly renormalize the step free energy, but it remains finite and positive [3, 4]. Anyhow, the reduction of the step free energy in the case of basal facet of ^{3}He crystal is more than two orders of magnitude which prevents the facet to appear at temperatures higher than 0.1 K [4] while the faceting/roughening transition is at 0.3 K.

The second statment, that is the prediction of the crystallization wave, is basically the notice that if the crystal can easily grow and melt under very small local pressure change, than the liquid can move like in the usual capillary waves with the surface tension of the deformed interface acting as the restoring force. The only difference with the capillary wave is that instead of the hump on the surface due to liquid fall, there will be a dip because of the growth of the crystal into the liq-

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uid, see Fig. 1. The amplitude of the solid bamp is large

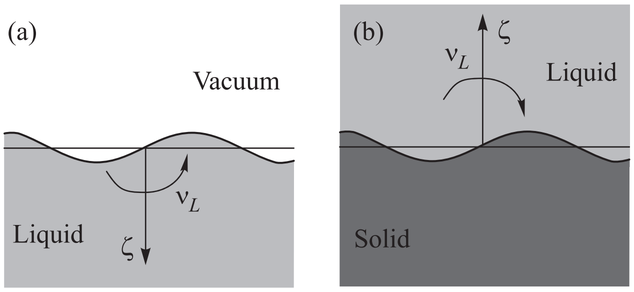

Fig. 1. (a) – Capillary wave on the liquid surface. (b) – Melting-freezing wave on the solid-liquid interface

than the amplitude of the dip in vacuum by factor of $\rho_L/(\rho_S - \rho_L)$ (about 10 for ^{4}He and 20 for ^{3}He) for the same velocity field in liquid, but this is basically all the difference between very well known capillary wave and the new exotic and counter-intuitive crystallization wave.

The close analogy between these two waves is clearly seen from the equilibrium condition at the interface. In usual capillary wave (Fig. 1a) the excess pressure in the liquid is compensated by the Laplace term, $\Delta P_L + \rho_L g\zeta = \alpha\zeta''_{xx}$ where $\zeta = \zeta(x, y)$ is the vertical distortion of the interface directed into liquid side and α is the surface tension. The velocity potential $\phi = \phi_0 \exp(iqx - i\omega t - qz)$ should satisfy the mass conservation condition at the interface $v_z|_0 = \partial\phi/\partial z|_0 = \dot{\zeta}$ which results in the well-known relation $\omega^2 = gq + (\alpha/\rho_L)q^3$.

In the case of solid-liquid interface we have similar force balance (Fig. 1b) $(\Delta P_S - \rho_S L g\zeta) - (\Delta P_L - \rho_L g\zeta) = -\gamma_x\zeta''_{xx}$ where instead of the surface tension α we have the surface stiffness $\gamma_x = \alpha + \alpha''_{xx}$ because the surface tension of the crystal is highly anisotropic. Co-existence of two phases implies the condition of equality of chemical potentials, $\Delta\mu_L = \Delta P_L/\rho_L = \Delta\mu_S = \Delta P_S/\rho_S$. The mass conservation condition transforms into $v_z|_0 = -(\Delta\rho/\rho_L)\dot{\zeta}$ which gives the spectrum

$$\omega^2 = g\frac{\rho_L}{\Delta\rho}q + \gamma\frac{\rho_L}{\Delta\rho^2}q^3. \qquad (1)$$

The relation (1) has been obtained by Andreev and Parshin in [2] 107 years after the spectrum of capillary waves was found by W. Thomson.

A year later the crystallization wave has been discovered in Kapitza Institute in Moscow by Parshin and his co-workers K. O. Keshishev and A. V. Babkin [5, 6]. They have used a capacitor made of 40 μm wires wounded bifilarly around a plane fiberglass holder to excite the wave by high electric field and they observed them through the set (five pairs) of optical windows. The excitation of the waves by an electric field is possible due to polarization of helium atoms which prefer to be in a high field region so that the more dense solid phase lifts up and oscillate if a high AC voltage is applied to the capacitor. To make the surface horizontal, a contact angle of about 135 degrees between the surface and the capacitor was tuned to 90 degrees by a DC voltage. The cryostat driven by evaporation of ^{3}He has allowed optical observations down to 0.36 K. In the same experiment the second, "a"-facet of ^{4}He crystal has been discovered [6]. The shape of the waves has been measured by reflection of laser light from the crystal surface. The measured spectrum of the crystallization waves was in the perfect agreement with the theoretical prediction (1), which is shown in Fig. 2.

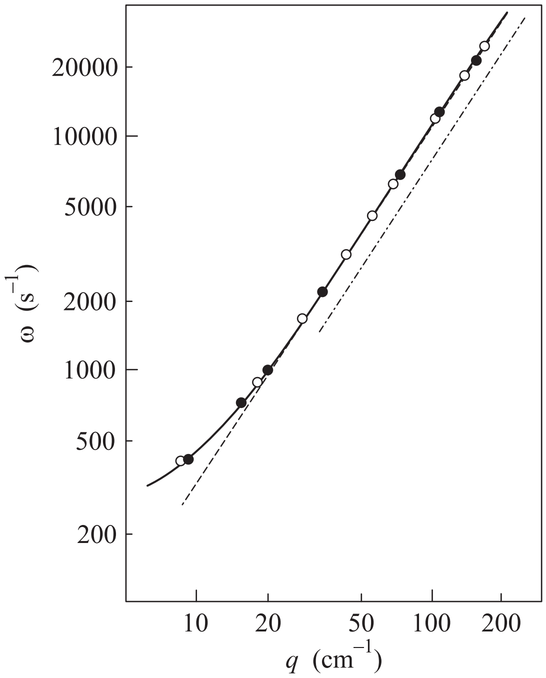

Fig. 2. The spectrum of crystallization waves as measured by Keshishev et al. [6]. The straight line indicates 3/2-power-law dependence

The damping of crystallization waves is essentially due to the finite mobility of the solid-liquid interface. When deriving (1) above we have assumed that the surface is always in equilibrium meaning that the mobility, or, growth coefficient $k \equiv \dot{\zeta}/(\Delta\mu_S - \Delta\mu_L)$ is infinitely high. If it is not, the dissipation term $(\rho_S/k)\dot{\zeta}$ must be added to the force balance, and Eq. (1) transforms to

$$\omega^2 = g\frac{\rho_L}{\Delta\rho}q + \gamma\frac{\rho_L}{\Delta\rho^2}q^3 - i\frac{\rho_S\rho_L}{\Delta\rho^2}\frac{\omega q}{k}. \qquad (2)$$

The dissipation of the waves has been also measured in this pioneering work [6] and has been shown to be determined by the collision of the moving interface with phonons and rotons. The Fig. 3 shows the phonon contribution proportional to T^4 and the remaining roton contribution.

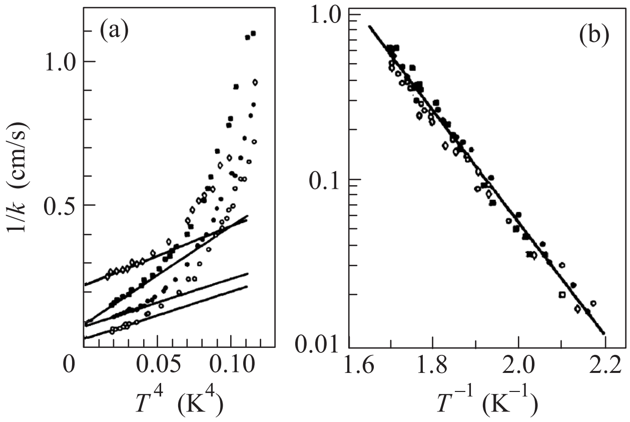

Fig. 3. The dissipation of crystallization waves as measured by Keshishev et al.: (a) – T^4 at low temperatures due to phonons. (b) – Dumping due to rotons after subtraction the phonon's contribution. The straight line corresponds to the roton activation energy of 7.8 K [6]

The dissipation has been measured at lower temperatures down to 40 mK by Wang and Agnolet [7]. They have used interdigital capacitors made by evaporation of metal onto borosilicate glass substrate in the shape of long thin fingers with the periodicity of 80 μm to excite the standing crystallization wave and to detect it. In their measurements the contact angle between the surface and the capacitor has been compensated by appropriate tilt of the capacitors, see Fig. 4a. They have observed a crossover from ballistic regime of collisions of moving interface with phonons to a hydrodinamic regime at about 0.25 K (Fig. 4b).

Crystallization wave has been proven to be a perfect tool to measure the highly anisotropic surface stiffness γ of the solid. Moscow group was first to measure the angular dependence of the stiffness [8], as shown in Fig. 5a. The experimental cell was able to change it's tilt by a large angle, so that the orientation of the grown crystal could be determined by observation of basic facets and then changed in the controlled way. As one can see, the stiffness at the orientations far from high symmetry directions is quite small, about 0.2 erg/cm^2, while at the orientations of basal "c" and "a" facets it is about 0.3–0.4 erg/cm^2. This information is of very high importance because the temperature of the transition from faceted

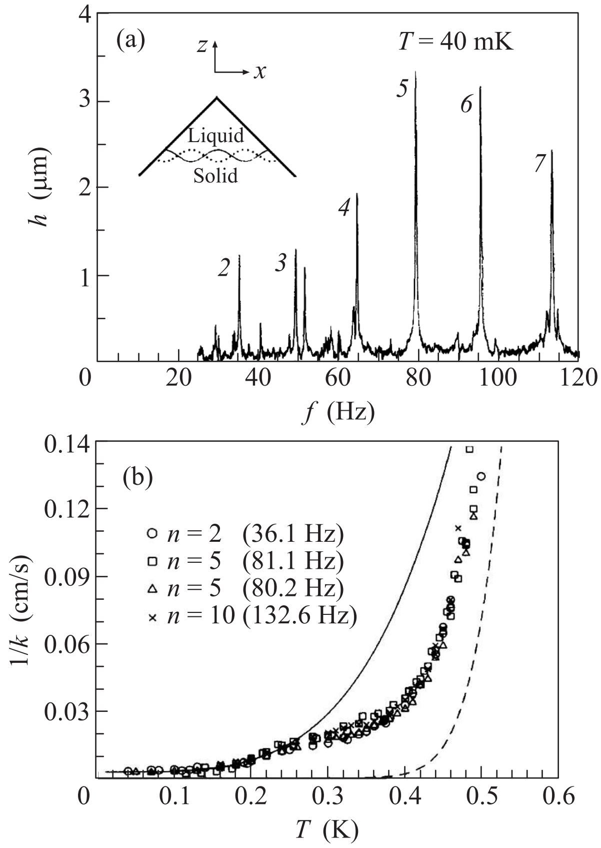

Fig. 4. (a) – The standing crystallization waves as observed by Wang and Agnolet [7]. (b) – The dissipation of crystallization waves: a clear crossover is seen at about 0.25 K from ballistic phonons to the hydrodinamical ones

to rough state is proportional to the surface stiffness, $T_R = (2/\pi)\gamma d^2$ where d is the interplanar distance.

Later the measurements of the surface stiffness have been continued by S. Balibar and co-workers in Paris [9]. In their measurements they have also used an optical cryostat with five pairs of windows thermally anchored at different temperatures to reduce the IR radiation heat leak. The minimum temperature of 40 mK of this experiment was by the order of magnitude lower compared to the one in Moscow experiments due to the use of the dilution of ^{3}He in liquid ^{4}He and due to a proper thermal shielding. In these experiments the interdigital capacitor similar to the capacitors used by Wang and Agnolet has been used to excite the waves and the laser light reflecting from the crystal surface has been used to measure the wave profile. The capacitor has been tilted to compensate the most of the contact angle which was then tuned accurately by the DC voltage to have perfectly horizontal surface. The angular resolution in Paris experiments was also extremely high which allowed Balibar's group to observe, for the first time, the predicted

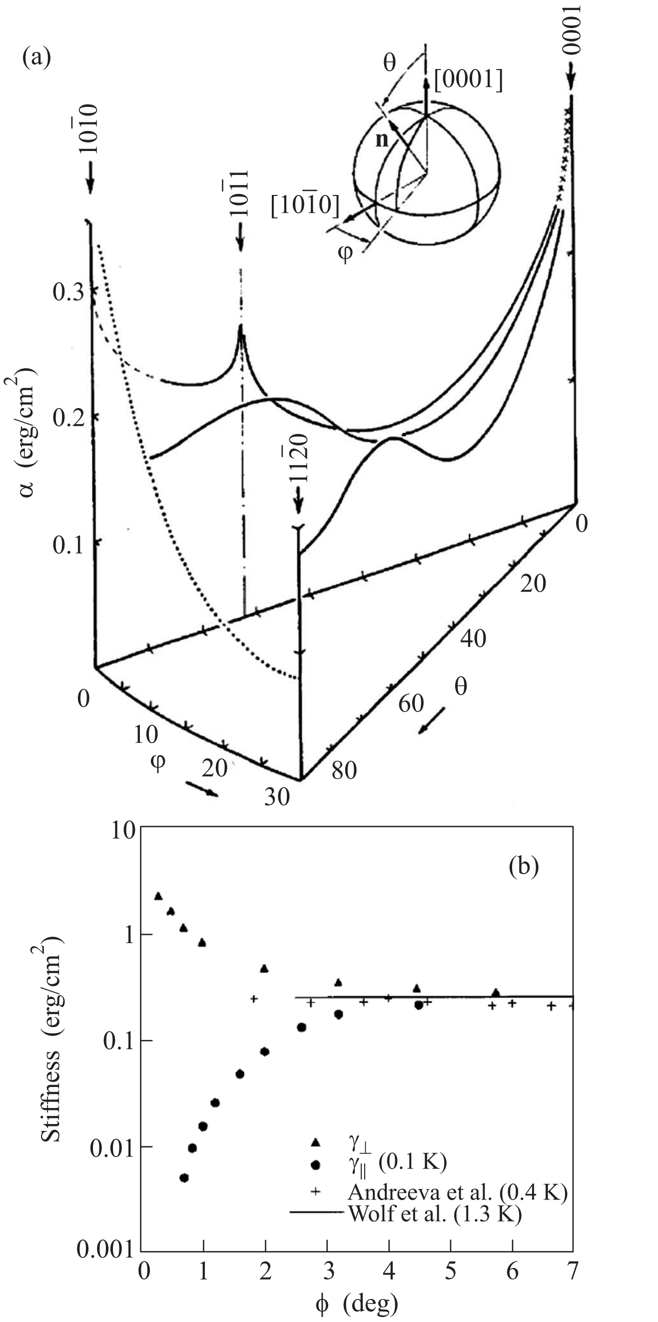

Fig. 5. (a) – The surface stiffness measured by Andreeva and Keshiishev [8]. (b) – The surface stiffness of vicinal surfaces as measured by Rolley et al. [9]

divergence of different components of the surface stiffness at the orientations close to the direction of basal c-facet.

The stiffness of the vicinal (close to the facet) orientation is due to the stiffness of and the interaction between elementary steps because the surface tilted by a small angle θ with respect to the facet can be viewed as the atomically flat terraces with the orientation of the facet separated by elementary steps with the density θ/d. The perpendicular component of the stiffness is thus due to the tension of individual steps β, $\gamma_\perp = \beta/(d\theta)$, while the parallel component is due to the repulsive interaction $\varepsilon = (\delta/d^2)\theta^2$ between neighboring steps, $\gamma_\perp = (6\delta/d^3)\theta$. The perpendicular component of the stiffness thus goes to zero when θ approaches zero and the perpendicular component goes to infinity which has been indeed observed in Paris experiments at the angles less than 3 degrees as shown in Fig. 5. The critical angle below which the surface behaves as vicinal one is determined by the width ξ of the elementary step: at angles higher than d/ξ individual steps overlap and the concept of terraces and steps fails. As we can see, the crystallization wave is a perfect tool to measure the surface stiffness locally with extremely high angular resolution to prove the basic concepts of the roughening transition and of the structure of vicinal surfaces.

In the year 2007 Gusev and Parshin [10] have demonstrated that the crystallization wave can also propagate along an atomically smooth surface, i. e. facet. The facet can grow and melt only by a motion of the elementary steps, and this process is much slower than the motion of a rough surface. Anyhow, Gusev, and Parshin have shown that a soliton-like wave in the form of the banch of the steps can exist and move over the facet. Unlike the wave on the rough surface, the wave on the facet is non-linear and it's spectrum differs much from the usual crystallization wave,

$$\omega^2 = \frac{8}{3\pi}\frac{\rho_L\gamma}{\Delta\rho^2}v^2\eta_0 q^4, \tag{3}$$

where η_0 is the amplitude of the wave and $v = \lambda/\sqrt{(32\pi/3)(\gamma\rho_L\eta_0/\Delta\rho^2V^2)}$. This wave has not yet been observed experimentally.

Up to now the crystallization waves have been seen only at the surface of ^{4}He crystals. The observation of the wave on the surface of ^{3}He srystal is much more difficult but very interesting. Due to the nuclear spin there appears an additional term in the external magnetic field H, which can be thought as the additional effective mass,

$$\omega^2 = \frac{\gamma q^2}{M},\ M(q) = \rho_L d_m\left(\frac{H}{H_0}\right)^2 + \frac{\Delta\rho^2}{\rho_L q}, \tag{4}$$

$$d_m = \frac{\chi_L c_{mL}^2}{\chi_S c_{mS}\Omega_S + \chi_L c_{mL}\Omega_L}, \tag{5}$$

where $H_0 = (c_{mL}/\chi_S)\sqrt{\rho_L\chi_L}$, c_m is the magnon velocity and χ is the magnetic susceptibility [11]. Such a magnetic surface wave would be extremely interesting to observe and investigate, but the damping of the crystallization wave in ^{3}He becomes reasonably low only at very low temperatures. First of all, liquid ^{3}He becomes superfluid only below 2.5 mK (to be compared with 2.1 K for ^{4}He), and the dissipation due to magnons in solid becomes low enough only at $0.1\,T_c \approx 0.2$ mK [12]. The temperature of $0.1\,T_c$ of ^{3}He is on the level of records in ultra low temperature experiments. Anyhow, the damping due to magnons is probably overestimated which have been shown by the measurements of mobility of ^{3}He crystal surface by Junnes et al. in Helsinki [13]. If one scales the measured growth coefficient of the rough surface of ^{3}He crystal as T^4, a temperature of 0.4 mK would be low enough for the observation of crystallization wave. The first attempt to see this wave in ^{3}He is currently in progress in O. V. Lounasmaa Lab in Helsinki [14]. Alexander Parshin has contributed a lot to this experiment and one can see that the crystallization wave phenomenon is very closely connected with him. He was the first to predict and observe the Parshin crystallization waves and he explored them till his death.

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