

Basic Research on Crystal Growth and Surface Physics of Solid Helium under Microgravity

Yuichi Okuda

Department of Condensed Matter Physics, Tokyo Institute of Technology
Oh-okayama, Meguro-ku, Tokyo 152-8551, Japan

Takao Mizusaki

Department of Physics, Graduate School of Science, Kyoto University
Kitashirakawa, Sakyo-ku, Kyoto 606-8502, Japan

Makio Uwaha

Department of Physics, Nagoya University
Furo-cho, Chikusa-ku, Nagoya 464-8602, Japan

1 Introduction

Liquid helium is regarded as a quantum fluid which is melted by the quantum fluctuation (zero point motion) even at $T = 0$. Liquid ^4He becomes superfluid at $T=2.17$ K at saturated vapor pressure, which is the manifestation of Bose-Einstein condensation, and liquid ^3He , which is a fermionic system, sets into the BCS-type superfluid at around 1 mK. The most outstanding feature of these quantum systems in the low temperature would be the fact that they are extremely pure. All foreign substances are frozen and excluded out of the system. And the thermodynamic parameters, such as temperature and pressure, to specify the system can be controlled very accurately and precise measurements can be performed. They are ideal systems to do a basic research about strongly interacting bosonic and fermionic systems.

If we apply an external pressure up to 25 bar for liquid ^4He and 35 bar for liquid ^3He , they are forced into the solid phase due to their strong repulsive interactions. Then we have nice quantum solids which coexist with the quantum fluids at very low temperature.

There should be a naive physics in the interface between the quantum solid and the quantum fluid. Here we would like to focus on the basic physics of crystal shape and crystal growth together with the unique new phenomena which are observed on the interface of the quantum solids.

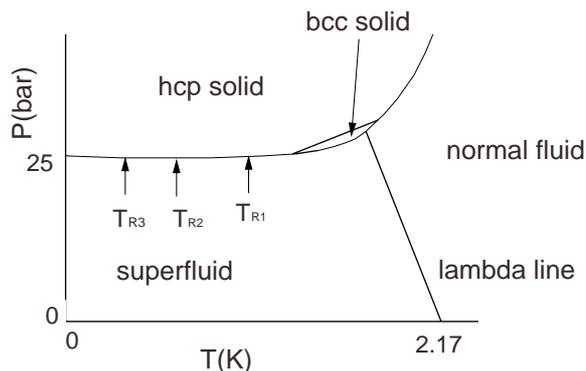


Figure 1: Phase diagram of ^4He in low temperature.

As can be seen from Fig.1, the melting curve is flat below 1 K. This means there is almost no latent heat involved in the liquid/solid transition, as is derived from the Clausius-Clapeyron relation. No heat diffusion problem comes in, which makes the crystal growth very difficult to analyze. And furthermore the melt of solid ^4He is superfluid and the mass transport proceeds very easily. Then the crystal growth of solid ^4He in low temperature can proceed with enormous speed. The growth velocity is proportional to the chemical potential difference and the coefficient, which is called as growth coefficient, is huge; something like 10^{10} times larger than the ordinary solid case. The crystal forms its equilibrium shape within a very short time. It can

be possible to obtain the microscopic information, such as the step-step interaction, from the direct observation of the crystal shape.

One of the spectacular examples of the interface of solid ^4He is that the crystal forms horizontal surface under the gravity to minimize the gravitational energy, similar to a liquid. And even more, if some perturbation is applied, the horizontal interface oscillates with small damping, which is called a crystallization-melting wave. This wave is a very unique wave only observable on the quantum solid surface in co-existence with superfluid.

Steps and kinks, which play essential roles in the physics of an interface, are regarded here as quasi-particles on the interface. Their numbers are fluctuating due to thermal and quantum fluctuation even at low temperature. A step can be a line-defect on the 2-dimensional plane and a kink be a point-defect on the 1-dimensional step. They are interesting quantum objects which are only realized on the surface of the quantum solids.

It is already realized that the ^4He interface is a new and ideal system to study any crystal-related physics. So far extensive results are accumulated [1] and the field is still progressing[2].

Present authors also contributed in this field; for example,

- (1) sound induced melting of solid[3],
- (2) negative crystal observed in solid ^4He [4],
- (3) crystal growth and melting of ^3He in U2D2 ordered solid[5],
- (4) quantum nucleation of solid[6, 7], and so on.

But the precise and thorough research will not be completed without the experiment under the microgravity.

2 Microgravity experiment relevance

As mentioned above, the very precise and basic measurement for crystal growth and surface physic is possible with the ^4He crystal, but unfortunately, it is found that the gravity obscures the experimental results.

The pressure gradient in gravity both in the liquid and the solid hampers the precise measurement of pressure. And not only the liquid compresses the solid to deform, but the crystal itself is easily deformed by its own weight. Such a deformation makes the true equilibrium shape obscure and makes the first principle of getting

the crystal shape (Wulff's theorem) not applicable.

Such gravity effect may be eliminated by the experiment with a tiny crystal. But it does not work out for the helium crystal. Due to a larger quantum fluctuation, the step on the solid He gets rather wide, though the concept of step is still meaningful. Owing to this effect, the facet size with respect to the total size of the crystal is very small compared with the classical solids, even if temperature is well below the roughening transition temperature. Then we do need a bigger crystal. The experiment with a bigger crystal is difficult on Earth because of the bigger gravity effect.

Another benefit to carry out experiment under microgravity is that the crystal can be studied, which is free from any contact with the wall. We can essentially access to all crystallographic orientations. This is totally impossible on earth.

3 Proposed objectives

We would like to propose the following experiments for both ^4He and ^3He . Chemically these species are similar, but in low temperature they behave quite differently due to the difference of the quantum effect. Of course ^4He is studied in more detail and easier to be studied, but the importance of the ^3He is clear. The quantum effect is larger than ^4He and ^3He carries a nuclear spin which should play an important role on the crystal growth. But crystal surface details are not well understood at the moment. Comparison between ^3He and ^4He results should be very instructive and important.

3.1 Equilibrium crystal shape

The transition region between a facet and an adjacent curved part of a crystal is known as a vicinal surface, which is shown in Fig. 2. The vicinal surface is only meaningful for the angle θ less than 3 deg, where steps are well separated by flat terraces. This surface is an ideal system to realize steps and kinks as quasi-particles on the surface and the growth rate of this surface is determined by the dynamics of those quasi-particles.

The functional form of this surface manifests the detailed step-step interaction. If the interaction is dominated by the d^{-2} term, the profile of

the surface $z(x)$ is proportional to $x^{3/2}$, where d is the distance between the steps. It is believed that this form holds for general crystals. But it is not yet confirmed experimentally. The stringent test of this theory can be done with the equilibrium crystal shape (ECS) of ^4He crystal. The observation of ECS with undeformed crystal is essential to determine the step-step interaction.

Experimental results on earth are different among different groups. The exponent of x is ranging between 1.2 and 1.8. The result is controversial. Main reason is that the crystal is deformed by gravity[8]. The rigorous test of the true functional form of the ^4He vicinal surface is only possible under microgravity.

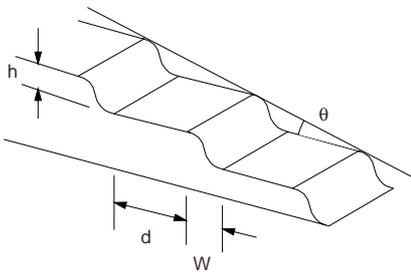


Figure 2: Vicinal surface inclined by a small angle of $\theta \ll 1$ with respect to a high symmetry facet. In helium crystal a step is not so sharp as the classical ones.

3.2 Roughening transition

It is regarded that roughening transition is classified as the Kosterlitz-Thouless (KT) transition, which is the transition of the 2-dimensional (2D) XY-type universality class. The following relation between the transition temperature, T_R and the stiffness of the surface at T_R is known.

$$kT_R = \frac{2}{\pi}\gamma h^2, \quad (1)$$

where h is the lattice period perpendicular to the surface, and γ is the stiffness of the surface.

Roughening transition was clearly observed on [0001] face of ^4He at $T_{R1}=1.23$ K and the rigorous test whether it is KT transition was done for the first time. The transition was identified by observing the curvature of that plane.

Following to the c-facet, other two facets (a-facet and s-facet) was observed at the transitions 0.9 K and 0.36 K, respectively. Though the

roughening transition of c-facet is rather clear, the other two facets were not observed in a equilibrium condition. They were observed only in the growing process.

There is an open issue in roughening transition of ^4He . In the recent experiment[9], ^4He crystal was cooled down to 2 mK, but no other roughening transition and no sign of freezing of kinks were observed down to that temperature. According to eq.(1), most low index planes have to be faceted at 2 mK.

The ground state of the crystal surface is expected to be the faceted state by classical theory. But for the quantum crystal, it is not clear whether it is faceted or rough at $T = 0$. Andreev and Parshin was the first to point out the possibility of the quantum roughening [10]. They argued quantum fluctuation may wash out the surface ordering even at $T=0$. Contrary to this, the renormalization theory [11] predicted that the crystal lattice periodicity eventually takes over whatever quantum fluctuation is. At least for the high Miller index planes, the roughening transition temperature is extremely low especially for solid helium. The concept of quantum roughening is a very important problem which must be tested by experiment under microgravity.

However, as long as the experiment is performed on Earth, the definite conclusion may not be obtained. If roughening transition is defined as the diverging point of the height-height correlation function, the gravity kills the transition. In real situation, the gravity will bring a smearing effect on the roughening transition. The roughening transition experiment should be done in space.

3.3 Instability of the quantum surface of solid

Since the solid-superfluid interface of He is very sensitive to an external perturbation, process of crystallization and melting propagates as a wave if the solid surface is not faceted. Surface tension is the restoring force to make the interface flat to recover the equilibrium shape from the perturbed shape. It is effective only at small wavelengths and the gravity is responsible for maintaining the flat interface at long wavelengths (this is why we observe a horizontal interface in He). Thus without gravity it is not possible to stabilize the solid-liquid interface against some external perturbations.

This is clearly seen in the dispersion of the

crystallization wave. When the solid encounters a non-hydrostatic uniaxial stress $\delta\sigma_{xx}^0$ the wave frequency is [12]

$$\omega_q^2 = \left[\alpha q^2 - 2q(\delta\sigma_{xx}^0)^2 \frac{1 - \sigma_P^2}{E} + (\rho_s - \rho_l)g \right] \frac{\rho_l}{(\rho_s - \rho_l)^2} q \quad (2)$$

where σ_P is the Poisson ratio and E the Young modulus. If $g = 0$, the frequency becomes imaginary, the wave fluctuation is amplified and the interface is unstable for a small wave number q . Similarly, another instability is predicted [13] if superfluid at the interface flows with a velocity v . The interface becomes unstable when the flow velocity exceeds the critical value

$$v_c = \left(4 \frac{g\alpha}{\rho_l} \frac{\rho_l}{\rho_s - \rho_l} \right)^{1/4}. \quad (3)$$

Without gravity the critical velocity vanishes, and any tangential flow at the surface makes the interface unstable.

The experimental implication is that under microgravity the surface of ^4He crystal is macroscopically unstable with a very weak uniaxial stress or very slow superfluid flow. In principle the former instability with stress is universal for all solids, but in practice is observable in a macroscopic scale only in solid ^4He which coexists with superfluid. The latter instability with flow, if observed, is very new phenomenon, in which superfluidity plays an essential role.

3.4 Crystal growth of nuclear-spin-ordered solid ^3He

There have not been much studies for the case of solid ^3He since liquid is the Fermi liquid (rather viscous liquid) and the entropy difference between solid and liquid stays constant to be $R \ln 2$ down to very low temperatures because of the paramagnetic nature of the nuclear spin in solid and Fermi liquid condensation in liquid ^3He . The growth rate for solid ^3He is extremely slow above 10 mK in comparison with ^4He , so that it sometimes takes many days to get to be in equilibrium. In order to set up the similar situation for ^3He as for ^4He , we have to cool the sample much below the nuclear-spin ordered temperature of the solid ($T_N = 0.93$ mK) where the latent heat of the solid/liquid is very small and the liquid is superfluid.

New features of the crystal growth of solid ^3He are that 1) quantum nature in ^3He is much stronger than in ^4He due to a smaller atomic

mass and 2) the crystal growth is governed by nuclear spins in solid and the transport of spins either in liquid or solid. One of spectacular predictions of ^3He is the existence of the magnetic crystallization wave below 0.1 mK[14]. Those unique natures of the ^3He crystal can be studied in detail only under microgravity as well as in the ^4He case.

Preliminary results of crystal growth and melting of nuclear-ordered solid ^3He was obtained down to 0.5 mK[5]. We found a large difference in growth and melting coefficients, that was attributed to the difference of crystal growth rates between a faceted surface in grow process and a rough surface in melting. We confirmed that the crystal growth (melt) was completely governed by nuclear spins and thus was sensitive to magnetic field and the nuclear spin structure.

4 Experimentals

4.1 Low temperature cryostat

For the experiment proposed above we need low temperature of 1 K-50 mK for a long period (one month). The dilution refrigerator is a common apparatus to get such low temperature, but it is too difficult to operate it in space. So we would like to develop so called a continuous adiabatic demagnetization refrigerator.

The idea is simple. The two sets of common adiabatic demagnetization stage is operated alternatively to get low temperature below 100 mK continuously. Key technology would be a cooling substance and heat switch technology. Chrome potassium alum salt and superconducting heat switch may be the best combination for the temperature.

The joint project on development of mK - refrigerator is planned with a domestic cryogenic group.

4.2 Acoustic microscope technology

Of course, optical measurement is essential for the experiment to monitor how the crystal is under microgravity. In addition to the optical measurement, we would like to propose to exploit the acoustic microscope technology to investigate the ^4He crystal surface.

Sound wave is a very useful tool to probe the elastic properties of the helium surface. Using a concave acoustic lens which is shown in Fig. 3 schematically, the sound wave can be focussed on the crystal surface at a very narrow spot. As the sound wave propagates in superfluid, we can

use a very high frequency without any dissipation. And the wave length could be as small as, or even smaller than, that of the visible light easily[15].

By scanning the lens in X-Y-Z directions, the reflected signal can construct a three-dimensional elastic image of the crystal. Using a tungsten tip nucleator of the crystal seed as shown in Fig. 3, we will be able to get the desired crystal orientation in the microgravity environment.

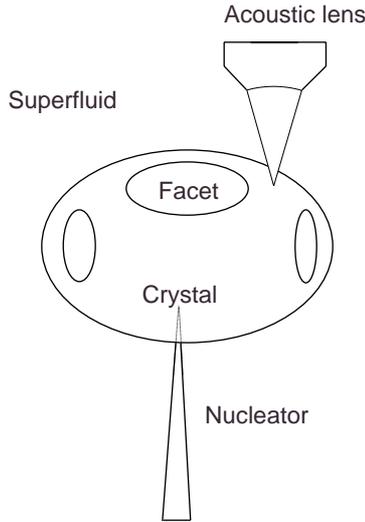


Figure 3: Schematic diagram of acoustic microscope investigating the vicinal surface, shape and elastic property.

4.3 Double demagnetization and MRI microscope

In order to carry the project of the crystal growth and surface physics in ^3He , we have to develop two technologies. One of them is the refrigerator operative under micro gravity, which is able to cool systems down to sub-mK temperature range. This would be possible to develop the double demagnetization refrigerator, where the 1st stage should be the reciprocally-operated electronic demagnetization stage as described in section 4.1 and the 2nd stage should be the nuclear demagnetization refrigerator. This refrigeration technology in space is a challenging one which Japanese research team should develop and offer to the world of low temperature community.

Second technique which is important to study the problem is the development of the nuclear magnetic resonance imaging (MRI) microscope. In this research, nuclear spin and its ordered

structure should be dominant factor for crystal grow and equilibrium shape of the crystal, it is essential to obtain a magnetic information of nuclear spin in space. We developed MRI microscope applicable at ultra-low temperatures [16, 17] and we had achieved the 2 dimensional space resolution of the MRI microscope to be 10 micron and the ultimate resolution could be improved to be in one macron size. We expect the MRI microscope will be a key tool to study crystal growth of ^3He and new physics will be explored in the crystal growth of ^3He under the microgravity.

5 Conclusion

The surface of the solid helium provides a new and unique system for the fundamental understanding of the crystal growth and surface physics. But the essential result will be obtained only if the proposed experiments are performed under microgravity. We have to overcome a few technical problems. But we believe it will open up a new field and contribute the fundamental condensed matter physics.

This kind of project should be proceeded with the international collaboration. We are already promoting the joint research with foreign groups.

Finally we would like to thank the cordial support from NASDA and Mitsubishi Research Institute for the present work.

References

- [1] S. Balibar, P. Nozieres, Solid State Commun. **92**, 19(1994).
- [2] M. Uwaha: Solid State Physics (Japanese) **20**, 431 (1985).
- [3] Y. Okuda, S. Yamazaki, T. Yoshida, Y. Fujii and K. Matsumoto, Physica **B 263-264**, 364 (1999).
- [4] Y. Okuda, S. Yamazaki, T. Yoshida, Y. Fujii and K. Matsumoto, J. Low Temp. Phys. **113**, 775(1998) .
- [5] R. Nomura, H.H. Hensley, T. Matsushita and T. Mizusaki J. Low Temp. Phys. **94**, 377-383 (1994).
- [6] Y. Sasaki and T. Mizusaki, J. Low Temp. Phys. **110**, 491 (1998).

- [7] S. Balibar, T. Mizusaki and Y. Sasaki, *Physica* **B284-288**, 248 (2000) and *J. Low Temp. Phys.*, to be published in September (2000).
- [8] J. E. Avron. R. K. P. Zia, *Phys. Rev.* **B37**, 6611(1988).
- [9] J. P. Ruutu, P. J. Hakonen, A. V. Babkin, A. Ya. Parshin and G. Tvalashvili, *J. Low Temp. Phys.* **112**, 117 (1998).
- [10] A. F. Andreev and A. Ya. Parshin, *Sov. Phys. JETP* **48**, 763(1978).
- [11] D. S. Fisher and J. D. Weeks : *Phys. Rev. Lett.***50**, 1077 (1983).
- [12] S. Balibar, D. O . Edwards and W. F. Saam *J. Low Temp. Phys.* **82**, 119-143 (1991).
- [13] M. Uwaha and P. Nozieres, *J. Phys. France* **47**, 263-271 (1986).
- [14] A. F. Andreev, *Czechoslovak J. Phys.* **46-S**, 3043 (1996).
- [15] K. Karaki, M. Suzuki and Y. Okuda, *J. Appl. Phys.* **67**, 1680(1990).
- [16] Y. Sasaki, T. Ueno, K, Nishitani, H. Nakai, M. Fujiwara, K. Fukuda and T. Mizusaki, *J. Low Temp. Phys.* **113**, 921-962 (1998).
- [17] T. Ueno, Y. Sasaki, K, Nishitani, H. Nakai, M. Fujiwara, K. Fukuda and T. Mizusaki, *J. Low Temp. Phys.* **113** ,1043-1048 (1998).