Hyperbolic roton and solid nucleation in superfluid ^{4}He

Tomoki Minoguchi [1],

Institute of Physics, Graduate School of Arts and Sciences, University of Tokyo, Komaba 3-8-1, Meguro-ku Tokyo 153-8902, Japan

Abstract

The solidification model for superfluid ^{4}He is reviewed, where the symmetry breaking order parameter η is appropriately defined and included in addition to the density change ξ. As a remarkable feature, the model explicitly shows that the instability to the solid is associated with the instability against the fluctuation of η, namely the softening of 'hyperbolic roton'. The rate W of solid nucleation is calculated based on the model. In contrast to ξ, η is non-conserved quantity, and then it leads the novel exponents in W near the spinodal pressure.

Key words: quantum nucleation; solid liquid transition; phase-field model ; superfluid ^{4}He

We shall first review the 'hyperbolic roton' model[1] for the solidification of superfluid ^{4}He under pressure.

The solidification is established by the density deformations in the superfluid. We adopt the sound Hamiltonian H_s for the quadratic deformations. We add nonlinear terms by hand to H_s to describe the instability to the finite wave amplitude state, i.e., the solid state. The total Hamiltonian is then $H = E_{kin} + W$, where

$$E_{kin} = \frac{m}{2\rho_1 V} \sum_{\boldsymbol{k}} \frac{|\delta\dot{\rho}_{\boldsymbol{k}}|^2}{k^2}, \quad (1)$$

$$W = \frac{m}{2\rho_1 V}\Big(\sum_{\boldsymbol{k}\in D_1} \frac{(\omega^{ph}_{\boldsymbol{k}})^2}{k^2} + \sum_{\boldsymbol{k}\in D_2} \frac{(\tilde{\omega}^{r}_{\boldsymbol{k}})^2}{k^2} \Big) |\delta\rho_{\boldsymbol{k}}|^2 - \frac{\tilde{b}}{3}\int d^3\boldsymbol{r}\,\delta\rho(\boldsymbol{r})^3 + \frac{\tilde{u}}{4}\int d^3\boldsymbol{r}\,\delta\rho(\boldsymbol{r})^4. \quad (2)$$

Here $\dot{A}$ denotes time derivative of A, m the atomic mass, V the volume of sample, $k \equiv |\boldsymbol{k}|$ and $\delta\rho(\boldsymbol{r}) \equiv \rho(\boldsymbol{r}) - \rho_1 = 1/V \sum_{\boldsymbol{k}} \delta\rho_{\boldsymbol{k}} \exp(i\boldsymbol{k}\cdot\boldsymbol{r})$ the number density deviation from the mean value ρ_1 at position $\boldsymbol{r}$.

First we shall concentrate on $P = P_m$, the melting pressure, and determine the phenomenological parameters such as $\tilde{b}$, $\tilde{u} > 0$ in W. Regarding W as the thermodynamic potential, or $T-\mu$ function, $W = 0$ both at the uniform superfluid and at the uniform solid state.

The domain D_1 is the photon domain where the energy spectrum is approximated by $\omega^{ph}_{\boldsymbol{k}} = \sqrt{c^2k^2 + \tilde{\lambda}k^4}$. The domain D_2 is the roton domain, but here we propose somehow novel form for the roton spectrum in the 'hyperbolic' fashion: $\tilde{\omega}^{r}_{\boldsymbol{k}} = c'\sqrt{(k-G)^2 + \Delta^2/(\hbar c')^2}$. This spectrum is approximated by $\Delta/\hbar + \hbar c'^2(k-G)^2/(2\Delta)$ near $k = G$, and by $c'|k-G|$ otherwise. G is the magnitude of the roton minimum wave vector. The direct observation of roton spectrum near $P = P_m$ is not far from this form, and suggests $c' = c$ [2]. The 'roton mass' is $\Delta/c'^2 = 0.11m$ at P_m, which is occasionally close to the known value $0.13m$. So we may safely use $\tilde{\omega^r}_{\boldsymbol{k}}$ instead of usual parabolic roton spectrum at P_m. Later we will show that this hyperbolic form for roton is naturally derived for general pressures.

The advantage to take $\tilde{\omega^r}_{\boldsymbol{k}}$ is that $(\tilde{\omega^r}_{\boldsymbol{k}})^2/k^2$ in D_2 has the same form as $(\omega^{ph}_{\boldsymbol{k}})^2/k^2$ in D_1. This results in the simple free energy functional (4) being symmetric in $\nabla\xi$ and $\nabla\eta$.

Since the superfluid crystallizes into hcp solid, we assume

[1] E-mail: mino@phys.c.u-tokyo.ac.jp

$$\delta\rho(\boldsymbol{r}) = \frac{1}{V}\delta\rho_0(r) + \frac{1}{V}\delta\rho_G(r)\sum_{n=1}^{M} e^{i\boldsymbol{k}_n\cdot\boldsymbol{r}} \qquad (3)$$

with $|\boldsymbol{k}_n| = G$. We take summation by $M = 6$ on a plane to realize the simple hexagonal reciprocal lattice. The angle-independent envelopes $\delta\rho_0(r)$ and $\delta\rho_G(r)$ are introduced for the homogeneous nucleation of solid.

Substituting (3) into (2), and taking the dimensionless parameters $\xi \equiv \delta\rho_0/(\rho_1 V)$ and $\eta \equiv \delta\rho_G/(\rho_1 V)$, we obtain $W = \int d^3\boldsymbol{r}\, w(\xi,\eta)$, where

$$w(\xi,\eta) = \varphi(\xi,\eta) + \lambda(\nabla\xi)^2 + \lambda'(\nabla\eta)^2 \qquad (4)$$

with $\lambda' \equiv mc'^2\rho_1/(2G^2)M$ and $\lambda \equiv m\rho_1/2\tilde{\lambda}$. $\varphi(\xi,\eta)$ is the polynomial up to the 4th order in ξ and η. Because of the simplification (3), the expansion coefficients in (4) are completely determined by a few experimental data at P_m with the conditions $\varphi(0,0) = \varphi(\Delta\xi,\Delta\eta) = (\partial/\partial\xi)\varphi(\Delta\xi,\Delta\eta) = (\partial/\partial\eta)\varphi(\Delta\xi,\Delta\eta) = 0$. Here $(\xi,\eta) = (0,0)$ and $(\Delta\xi,\Delta\eta)$ are the values of the superfluid and the solid respectively. Especially, the characteristic energy density $P_0 \equiv mc^2\rho_1/2 = 115$bar, the smallness parameter $\varepsilon \equiv \Delta/(\hbar c' G) = 0.13$, the dimensionless coefficients $b \equiv \tilde{b}\rho_1^3/P_0 = 0.065$, $u \equiv \tilde{u}\rho_1^4/P_0 = 0.0038$.

It is worthy to note that $(\lambda',\Delta\eta)$ are derived to be $(1.21[K/\mathring{A}], 0.73)$, while $(\lambda,\Delta\xi) = (-3.60[K/\mathring{A}], 0.094)$ in a direct measurement. Then $|\lambda(\Delta\xi)^2| << |\lambda'(\Delta\eta)^2|$. This solves the paradox that $\lambda < 0$ may lead to the negative surface energy σ at the solid-liquid interface[3]: σ becomes positive here because of the presence of the energy of $\nabla\eta$ and gives a reasonable value. From now on, we safely drop $\lambda(\nabla\xi)^2$ from (4).

At pressures out of P_m, by putting $\Delta P \equiv P - P_m$, the density functional changes to $\varphi(\xi,\eta;\Delta P) \simeq \varphi(\xi,\eta) - \Delta P\xi + O(\xi^2)$. For all the pressures in question, we found that the higher order terms in ξ are negligible, and obtain φ in the form: $\varphi/P_0 = (\xi - D(\eta;\Delta P))^2 + E(\eta;\Delta P)$. That is, $\varphi(\xi,\eta;\Delta P)$ is parabolic increasing with ξ, and then the instability to the solid state does not occur no matter how the sound amplitude in D_1 is large under $\eta = 0$. The 'most probable' path (MPP) $\xi = D(\eta;\Delta P)$ ($\equiv \langle\xi\rangle$) connects the superfluid state $(\xi,\eta) = (\xi_1, 0)$ to the solid state $(\xi_1 + \Delta\xi, \Delta\eta)$ passing over the saddle point. Here $\xi_1 \equiv \Delta P/(2P_0)$. (Remember ρ_1 in scaling ξ is the mean liquid density *at* P_m. Then ξ_1 is finite at $P \neq P_m$.) If the solidification occurs quasi-statically, it does along the MPP. For simplicity, we shall assume that the solidification always goes along the MPP even when it does kinetically.

On the MPP, expanding (4) up to, say, the 3rd order in η, and neglecting constant, we have

$$w(\langle\xi\rangle,\eta) \simeq M(\varepsilon^2 - b\frac{\Delta P}{2P_0})\eta^2 + \frac{2}{3}Mb\eta^3 + \lambda'(\nabla\eta)^2. \qquad (5)$$

This means that the energy spectrum in D_2 changes to $\tilde{\omega^r}_{\boldsymbol{k}}(P)$ defined by $(\tilde{\omega^r}_{\boldsymbol{k}}(P))^2/k^2 \equiv (\tilde{\omega^r}_{\boldsymbol{k}})^2/k^2 - b\Delta P/(2P_0)$, and then

$$\tilde{\omega^r}_{\boldsymbol{k}}(P) = c'\sqrt{(k-G)^2 + \frac{\Delta(P)^2}{\hbar^2 c'^2}}. \qquad (6)$$

where $\Delta(P) \equiv \Delta\sqrt{1 - \alpha\Delta P/P_0}$ with $\alpha \equiv b/(2\varepsilon^2) \simeq 1.9$ and the 'roton mass' is $\Delta(P)/c'^2$. The coincidence of these roton parameters with experiment are satisfactory from 0bar to P_m (!).

(5) predicts that the superfluid becomes unstable at the spinodal pressure $P_c \equiv P_m + 2\varepsilon^2 P_0/b \approx 85$bar where the roton gap vanishes. Near P_c, we adopt (5) for the solid nucleation. By using $\eta_0 \equiv (P_c - P)/(2P_0)$ and $R_0 \equiv \sqrt{\lambda'/(P_0\eta_0)}$, we scale the parameters $\chi \equiv \eta/\eta_0$ and $\boldsymbol{x} \equiv \boldsymbol{r}/R_0$. Finally,

$$W = W_0 \int d^3\boldsymbol{x} \;\; (\;\frac{1}{2}\chi^2 - \frac{1}{3}\chi^3 + (\;\frac{\partial\chi}{\partial\boldsymbol{x}}\;)^2), \qquad (7)$$

where the pressure dependence is contained only in $W_0 \equiv b'\eta_0^3 R_0^3 P_0 \propto (P_c - P)^{3/2}$. The thermal nucleation rate is then $W_{th} \propto \exp\{-(P_c - P)^{3/2}\}$.

In this spinodal limit, we have

$$E_{kin} = \frac{m}{2G^2}\rho_1 M \int d^3\boldsymbol{r}\; \dot{\eta}^2, \qquad (8)$$

and then, using the scaled time $\tau = t/t_0$ with $t_0 \equiv \sqrt{m\rho_1/(4bG^2\eta_0 P_0)} \propto (P_c - P)^{-1/2}$, the action $S = \int dt(E_{kin} + W)$ is

$$S = S_0 \int d\tau d^3\boldsymbol{x}[(\frac{\partial\chi}{\partial\tau})^2 + (\frac{1}{2}\chi^2 - \frac{1}{3}\chi^3 + (\frac{\partial\chi}{\partial\boldsymbol{x}})^2)] \qquad (9)$$

with $S_0 = W_0 t_0 \propto P_c - P$. Then the quantum nucleation rate is $W_q \propto \exp\{-(P_c - P)\}$.

These exponents in W_{th} and W_q are consistent with experiment [4], [5] where the nucleation occurs on the wall of container. Although it is dangerous to compare our homogeneous result with these heterogeneous ones, it is quite attractive to study these connections.

We thank Dr. Seishi Kondo for collaborating at the initial stage. This research was supported by the Grant-in-Aid for Scientific Research (C), 11640337, 1999.

References

[1] T. Minoguchi, J. of Low Temp. Phys. **126** (2002) 627.

[2] O. W. Dietrich, E. H. Graf, C. H. Huang and L. Passell, Phys. Rev. A**5** (1972), 1377.

[3] Q. Xiong and H. J. Maris, J. of Low Temp. Phys. **77** (1989) 347.

[4] J.P. Ruutu, P.J. Hakonen, J.S. Penttila, A.V. Babkin, J.P. Saramaki and E.B. Sonin, Phys. Rev. Lett. **77** (1996) 2514.

[5] Y. Sasaki and T. Mizusaki, J. of Low Temp. Phys. **110** (1998) 491.