Quantum Nucleation on a Wall

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Abstract

Quantum nucleation of solid ^{4}He on a wall was discussed on a phenomenological model which describes incomplete wetting. While this model behaves as well-known thin wall nucleation model near the equilibrium point, it describes the nucleation as softening of ripplon on the adsorbed film near the critical point. As the corresponding experimental researches were done near the critical point, the nucleation rate W of the model was estimated in the the manner, which was developed by Lifshitz and Kagan, near the critical point. The result indicates the pressure dependence $\ln W \propto (P_{\rm c} - P)^{5/8}$, where $P_{\rm c}$ is a critical pressure. The assumption that the origin of metastability is the structural mismatch between the adsorbed film and the bulk leads to the value of $P_{\rm c}$.

Key words: quantum nucleation ; solid helium4 ;wetting

In the last few years, the study of quantum solid nucleation from superfluid ^{4}He has great interest among other studies of Macroscopic Quantum Nucleation(QMN). Because although other studies of MQN is concerning with isotropic process, it will occur on the wall of the container.

The experimental studies[1][2] of the system have reported temperature independent nucleation in the copper cell below 0.5-1 K. When P = 3-30 mbars above the melting pressure $P_{\rm m}$, the nucleation rate W indicates the pressure dependence

$$\ln W \propto (P_{\rm c} - P)^{\gamma}, \qquad (1)$$

where $P_{\rm c}$ is a critical pressure which corresponds to the instability point, and $\gamma \approx 1$. As for theoretical aspects, Balibar *et al.*[3] pointed out that a remnant crystal in the cavity of the wall has too high energy barrier if the value of $P_{\rm c}$ of this model were fitted on the experimental one. Uwaha[4] discussed the instability of film on the wall two decades before. But analysis was done near$P = P_{\rm m}$ and the discussion near $P = P_{\rm c}$ have never been tried. The aim of this paper to try it.

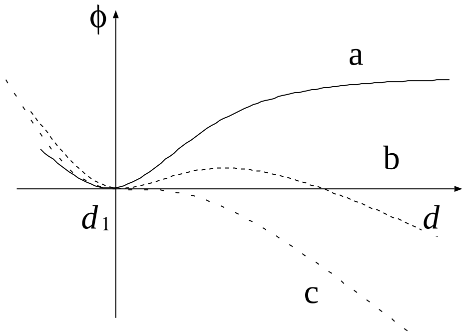

Fig. 1. Thermodynamic potential density classified in class-II against film thickness d. d_1 denotes the thickness of metastable state. Curve (a) and (c) correspond to $P = P_{\rm m}$ and $P = P_{\rm c}$ respectively. Curve (b) is the intermediate case between (a) and (c).

Dash[5] and Peierls[6] developed the phenomenological theory to treat the instability of flat film. They introduced Helmholtz free energy per unit area $\mathcal{F}$ as continual function of film thickness d and classified adsorption into three classes by the form of $\mathcal{F}$. In this paper, we construct our model based on the class-II category which corresponds to the incomplete wetting.

Here we define the potential energy of nucleation:

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$$U = \int \mathrm{d}S \left\{ \frac{1}{2}\sigma_{\mathrm{ls}} \left(\nabla_{\perp} d\right)^2 + \phi(d, d_1) \right\}, \qquad (2)$$

where $\nabla_{\perp} = (\partial/\partial x, \partial/\partial y)$, $\sigma_{\mathrm{ls}}$is the interfacial tension between liquid and solid, and S is the area of flat wall plane. ϕ is the thermodynamic potential density constructed by $\mathcal{F}$. On $P = P_{\mathrm{m}}$, $\delta U = 0$ reproduces the Young relation and near $P = P_{\mathrm{c}}$, U becomes the potential energy of nucleation discussed by Uwaha(see Fig.1(a)). The above two points are arguments that this model is reliable generic one. We analyze this model near P_{c}, (see Fig.1(c)) in the manner developed by Lifshitz and Kagan[7]. Using the Gibbs Duhem relation, the dimensionless parameter that decides the stability of the system becomes as follow:

$$\xi_0 \propto (P_{\mathrm{c}} - P)^{1/2}. \qquad (3)$$

At high temperature region, the Arrhenius low gives the nucleation rate. Therefore the dimensional analysis of U determines the pressure dependence of $\ln W$.

$$U = U_0 J\{\chi\}, \qquad (4)$$

$$J\{\chi\} = \int \mathrm{d}^2 x \left\{ \frac{1}{2} \left(\frac{\partial}{\partial \mathbf{x}} \chi \right)^2 + \chi^2 (1 - \chi) \right\}, \qquad (5)$$

$$\ln W = -\frac{U_0 J\{\chi\}}{k_{\mathrm{B}} T} \propto \xi_0^2 \propto (P_{\mathrm{c}} - P)^1, \quad \therefore\ \gamma = 1. \qquad (6)$$

Here, U_0 is the typical energy scale of the system and $J\{\chi\}$ is the dimensionless potential. At low temperature, thermal process of nucleation vanishes and quantum process becomes dominant. Next, we introduce the inertial term K to discuss the quantum process. The flow around the nucleus on the film leads to

$$K = \frac{m}{4\pi} \rho_{\mathrm{eff}} \int \mathrm{d}S \int \mathrm{d}S' \frac{\dot{d}(\mathbf{r}) \dot{d}(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}. \qquad (7)$$

Harmonic approximation of the Lagrangian $L = K - U$ gives the ripplon. From this viewpoint one may say that this nucleation is the softening of ripplon. If the dimensionless effective Planck constant $\hbar_{\mathrm{eff}}$ determined by U and K is enough small for WKB approximation,

$$\ln W = -\frac{s(\tau)}{\hbar_{\mathrm{eff}}} \propto (P_{\mathrm{c}} - P)^{5/8}, \quad \therefore\ \gamma = \frac{5}{8}. \qquad (8)$$

Here, $s(\tau)$ is dimensionless action of inverted potential $-U$. As the above mentioned, the values of γ in each process are not so far from experimental results. It is difficult to discuss discrepancy of the value of γ between these theoretical results and experimental results. We may say this model gives a good account of a aspect of the experimental results.

Although the above discussion is extremely general, it could not lead to the value of P_{c}. To estimate it, we have to consider explicit form of $\mathcal{F}$. Since the van der Waals potential makes the film stable, the mechanism to make film unstable is needed. The lattice spacing of adsorbed film on a wall may be somehow different from the bulk solid, because atomic structure of wall affect the film. Huse[8] considered the structural mismatch between the adsorbed film and bulk solid, and introduced film elastic energy parallel to the plane in phenomenological manner to discuss incomplete wetting. Applying his theory to ours, we rewrite $\mathcal{F}$:

$$\mathcal{F} = \alpha_{\infty} + \frac{\Gamma}{d_{\mathrm{m}}^2} \left(-\frac{d_{\mathrm{m}}}{d} + \frac{1}{2} \left(\frac{d_{\mathrm{m}}}{d} \right)^2 \right) + (\rho_{\mathrm{s}} - \rho_{\mathrm{l}}) \mu_{\mathrm{m}} d, \qquad (9)$$

where $\alpha_{\infty} = \sigma_{\mathrm{ws}} + \sigma_{\mathrm{sl}}$, d_{m} is film thickness on P_{m}, and Γ is the strength of the van der Waals potential. Using the above $\mathcal{F}$ and the Young relation,

$$\sigma_{\mathrm{ls}} (1 - \cos \theta_{\mathrm{cy}}) = \frac{\Gamma}{2 d_{\mathrm{m}}^2}. \qquad (10)$$

Here, θ_{cy} is Young's static contact angle. Moreover,

$$P_{\mathrm{c}} - P_{\mathrm{m}} = \frac{4}{27} \rho_{\mathrm{L}} \frac{\Gamma}{d_{\mathrm{m}}^3}. \qquad (11)$$

(10) and (11) lead to the conclusion that P_{c} is increasing for θ_{cy}.

These relations (10) and (11) are important to argue that this model explains the experimental results. We have to predict the value of P_{c} using them, but I left the problem untouched.

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