Quantum Nucleation in Helium under High-Frequency Field

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(Received January 14, 1994)

We study the kinetics of nucleating droplets in metastable helium at ultralow temperatures. Our theory takes into account intense acoustic or electric field. The quantum nucleation rate can be greatly enhanced by the field when its frequency ω is higher than the inverse of tunnelling time τ₁. In particular, we present a new formula for the quantum nucleation rate in the limit ω τ₁ ≫ 1.

quantum nucleation, ⁴He and ³He–⁴He, ultrasound, microwave

Recently, considerable attention has been paid to nucleation in superfluids ⁴He and ³He–⁴He at ultralow temperatures at which dissipative effects are very weak. As an original theory on metastable ⁴He, Lifshitz and Kagan proposed that the radius R of a droplet of the new phase is a semi-macroscopic quantum variable and that emergence of critical nuclei may be described as a quantum tunnelling phenomenon.¹) Subsequently Lifshitz et al. showed that the ³He–⁴He case can be treated using the same theoretical scheme.²) In accord with their prediction, recent experiments on supersaturated ³He–⁴He mixtures have shown that the supersaturation at which phase separation is triggered becomes almost independent of the temperature T below about 10 mK.³⁻⁵) However, the experiments have not yet been well explained and further theoretical effort is required.

In this letter, we present a new derivation of the kinetic equation for R in ⁴He neglecting dissipation, because the original derivation¹) is incomplete. Our final result coincides with the original one; hence, the details will be deferred to the Appendix. Then we examine effects of high-frequency electric or acoustic field (around 10 GHz, for example) on the quantum tunnelling rate. It can be greatly enhanced when the frequency ω is much higher than the inverse of a (classical) tunnelling time τ₁.⁶) Our formulae are applicable both to ⁴He and ³He–⁴He. Experiments, which will be proposed in this letter, seem to be very difficult in ⁴He but might be possible in ³He–⁴He.

The droplet radius R is assumed to be governed by the following Hamiltonian,¹,²)

\[ H = 2\pi \rho_{\text{eff}} R^3 \dot{R}^2 + 4\pi \sigma R^2 - \frac{4\pi}{3} \mu_{\text{eff}} R^3, \]  

(1)

where \( \rho_{\text{eff}} \) is a mass density, \( \dot{R} = \partial R / \partial t \), \( \sigma \) is the surface tension, and \( \mu_{\text{eff}} \) is chemical potential per unit volume. The first term is the kinetic energy due to the liquid around the droplet, while the last two terms are the usual terms in nucleation theory representing the energy of creating a droplet with radius R. If \( \mu_{\text{eff}} \) is slightly positive, the liquid is metastable and the critical radius \( R_c \) is given by

\[ R_c = 2\sigma / \mu_{\text{eff}}. \]  

(2)

Without dissipation, \( H \) is a constant of motion in the growth process, so that \( \dot{R} \) tends to a terminal velocity \( v_\infty \) for \( R \gg R_c \),

\[ v_\infty = \left[ 2 \mu_{\text{eff}} / 3 \rho_{\text{eff}} \right]^{1/2}, \]  

(3)

which is assumed to be much smaller than the velocity of sound. Supposing a droplet of solid ⁴He in a metastable superfluid ⁴He and neglecting the crystal anisotropy in the solid region, Lifshitz and Kagan found that

\[ \rho_{\text{eff}} = (\rho_s - \rho_l)^2 / \rho_l \]  

(4)

\[ \mu_{\text{eff}} = (\rho_s / \rho_l - 1)(p - p_{\text{cx}}), \]  

(5)

where \( \rho_s \) and \( \rho_l \) are the mass densities of the solid and the liquid, respectively, \( p \) is the pressure in the metastable liquid, and \( p_{\text{cx}} \) is the
pressure (\(\sim 25\) atm) of equilibrium two-phase coexistence.

When a sound wave is present, the pressure deviation \(\delta p = p - p_{ex}\) consists of a constant (dc) part and an oscillating (ac) part as \(\delta p = \delta p_0 + p_1 \cos (\omega x t)\). We may also apply an ac electric field \(E(t) = E_0 \cos (\omega x t)\) to a metastable helium system. Because the dielectric constant \(\varepsilon\) within a droplet is slightly different from that \(\varepsilon_0\) in the surrounding medium, a dipolar field is induced around a droplet under \(E(t)\). The disturbance changes on the spatial scale of \(R\), so that the excess energy \(\Delta H\) is proportional to \(R^3 E(t)^2 = 1/(2 R^3 E_0^2 [1 + \cos (2 \omega x t)]\). Because \(\Delta H \propto R^3\), \(\Delta H\) only changes \(\mu_{eff}\) in eq. (1). Therefore, both in acoustic and electric fields, \(\mu_{eff}\) may be expressed as

\[
\mu_{eff} = \bar{\mu}_{eff} [1 + \varepsilon \cos (\omega x t)].
\]  

To correctly calculate \(\Delta H\) or the parameter \(\varepsilon\) in eq. (6), we must introduce an \(E^2\)-dependent chemical potential and take account of the density and frequency dependence of the dielectric constant.\(^7\) This is a task for future study.

It is now convenient to introduce a new dimensionless variable,

\[
x = (2 R / 3 R_c)^{3/2},
\]  

where \(R_c\) is the critical radius in eq. (2) for the dc chemical potential \(\bar{\mu}_{eff}\). It should not be confused with the space coordinate. The time \(t\) will be scaled by \(\tau_0 = (3/5)(3 \rho_{eff} / 2 \sigma)^{1/2} R_0^{3/2}\), \(\omega\) by \(\tau_0^{-1}\), and the energy by \(E_0 = 9 \pi R_c^2\). Then we obtain

\[
H^* = H / E_0 = \frac{1}{2} x^2 + x^{4/5} - x^{6/5} [1 + \varepsilon \cos (\omega x t)].
\]  

In quantum mechanics \(\hat{x}\) in eq. (8) should be replaced by \(-i\hbar_{eff} \partial / \partial x\), where \(\hbar_{eff}\) is a scaled Planck’s constant,

\[
\hbar_{eff} = \hbar / (\tau_0 E_0) = (5h / 8 \pi)(\rho_{eff} \sigma)^{-1/2} \left( \frac{3}{2} \frac{R_c}{R_0} \right)^{-7/2}.
\]  

For realistic values of \(R_c (\sim 10 \AA)\), \(\hbar_{eff}\) remains extremely small. Let us consider the ground-state wave function for the Hamiltonian \((-1/2)\hbar_{eff}^2 (\partial / \partial x)^2 + x^{4/5}\) in which the chemical potential part is neglected. Its width is on order of \(x_Q = \hbar_{eff}^{3/7}\) and is much smaller than any characteristic values of \(x\) appearing in the following WKB calculation. In other words, most quantum fluctuations of \(R\) are on order of \(R_0 = x_Q^{15/8} R_c \sim (5h / 8 \pi \sqrt{\rho_{eff} \sigma})^{3/7} (\sim 1 \AA)\) and are much shorter than the exceptionally large quantum fluctuations needed to produce a critical droplet. Of course eq. (1) may not hold at small \(R\), because the droplet scheme, on which eq. (1) is based, can be used only when \(R\) is longer than the interface thickness.

The Schrödinger equation is

\[
i \hbar_{eff} \frac{\partial}{\partial t} \Psi = H^* \Psi,
\]  

in dimensionless form and the WKB equation for the wave function \(\Psi = \exp [i S(x, t) / \hbar_{eff}]\) is

\[
\dot{S} - \frac{1}{2} S'^2 + U_0 + U_1 \cos (\omega x t) = 0,
\]  

where \(\dot{S} = \partial S / \partial t, S' = \partial S / \partial x, U_0 = x^{4/5} - x^{6/5}\), and \(U_1 = - \varepsilon x^{6/5}\). If \(U_1\) is neglected, we obtain the zeroth-order solution

\[
S_0 = i \int_0^x dx_1 [2 U_0(x_1)]^{1/2}.
\]  

The deviation \(S = S - S_0\) obeys

\[
\dot{S}_1 + i (2 U_0)^{1/2} S_1^2 + \frac{1}{2} S_1^2 + U_1 \cos (\omega x t) = 0.
\]  

We can integrate eq. (13) by neglecting the nonlinear term \((1/2)S_1^2\) at small \(x\). We impose the boundary condition \(S = 0\) at \(x = 0\) and introduce a penetration time \(\tau(x)\) at the position \(x\),

\[
\tau(x) = \int_0^x dx_1 [2 U_0(x_1)]^{-1/2}.
\]  

Here \(\tau(x) \propto x^{3/5}\) for \(x \ll 1\) and \(\tau = \tau(1) = 5 \pi / 4 \sqrt{2} \approx 2.8\) is the tunnelling time in dimensionless units. We write the linearized version of eq. (13) as

\[
\left( \frac{\partial}{\partial \tau} + i \frac{\partial}{\partial \tau} \right) S_1 + U_1 \cos (\omega x t) \approx 0.
\]  

Because \(\partial / \partial \tau + i \partial / \partial \tau = i(\partial / \partial \xi)\) with \(\xi = t + i \tau\) held constant, we obtain
where $x_1$ is the integration variable and $\tau_1 = \tau(x)$. It is important that, for $\omega \tau \sim \omega x^{3/5} \gg 1$, or for $x > x_{\omega} = \omega^{-5/3}$, $S_1$ grows exponentially with increasing $x$ as

$$S_1 \equiv -i e A_1 \alpha^{-3} \exp \left[ -i \omega t + \omega \tau(x) \right],$$

where $A_1 = 18/25 \pi \beta$. We may confirm the following for $\omega \tau_1 \geq 1$: (i) $S_1$ increases in the form of eq. (17) in the narrow region $0 < x < x_{\omega} = \omega^{-5/3}$. The oscillating potential is crucial only in this region giving rise to the coefficient of the exponential factor in eq. (17). (ii) For $x > x_{\omega}$, the balance $(\partial / \partial t + i \partial / \partial \tau) S_1 \equiv (-i \omega + i \alpha / \partial \tau) S_1 = 0$ holds in eq. (15) and there is a sizable region of $x$ in which the oscillating potential can be neglected. (iii) With further increase in $x$, the nonlinear term $(1/2) S_1^3$ becomes important while the oscillating potential is still negligible.

As a result, to linear order in $U_1$ in calculating $S$, the nucleation rate $J$ (the time average of $|\Psi|^2$ at $x = 1$) may be expressed for $\omega \tau_1 \geq 1$ as

$$J = J_0 \exp \left[ -\left( \frac{25 \pi}{64 \sqrt{2} \hbar} \right) \{ 1 - A_2 \alpha^3 (\omega \tau_1)^{-3} e^{\omega \tau_1} \} \right],$$

where $J_0 \sim e^{80}$ is a microscopic constant and $A_2 = 9 \pi^2 / 5 \equiv 17.8$. We note that $J$ coincides with the original expression by Lifshitz and Kagan for $\alpha = 0$ and that the oscillating perturbation becomes appreciable for $\omega \tau_1 \gg 1$ due to the exponential factor $\exp (\omega \tau_1)$ however small $\alpha$ is. Ivel and Mel’nikov have generally discussed similar exponential enhancement of the effect of high-frequency electric field on the tunnelling rate.

As $\omega \tau_1 \rightarrow \infty$, however, eq. (18) eventually becomes invalid. This high-frequency limit may be examined if the wave function $\Psi$ is expanded as

$$\Psi = \sum_{n=0}^{\infty} \frac{1}{n!} \left[ e A_1 / \omega^3 \hbar \right]^n \exp \left[ -i n \omega t - \frac{1}{\hbar} \int_0^x dx_1 \sqrt{2 U_0(x_1) - 2 \hbar \omega n} \right].$$

This form is valid for $\omega \tau(x) \geq 1$ or $x \geq x_{\omega}$ because eq. (17) has been used. In fact, if the exponent of the last factor is expanded in powers of $\hbar \omega$, $S_0$ and $S_1$ given by eqs. (12) and (17) may be reproduced. Equation (19) is a superposition of excited states with $E_n = n \hbar \omega$, and the low-energy states with $\hbar \omega \ll U_0(x)$ give rise to eq. (18). Here we notice that, if $E_n$ exceeds the maximum $U_{\max}$ of the potential $U_0(x)$, such high-energy states are not damped by the potential barrier as $R$ is expanded up to $R_c$ from the size $R_c/(\omega \tau_1)^{2/3}$ at which $\omega \tau(x) \sim 1$. Summing up the terms with $n > U_{\max} / \hbar \omega$ in eq. (19), we find another nucleation rate from these highly excited states,

$$J' = J_0 \exp \left[ -\frac{4}{\hbar \omega} U_{\max} \ln (\omega \tau_1 / A_3 \alpha^{1/2}) \right],$$

where $J_0'$ is a microscopic quantity and $A_3 = 9 \pi (3 \alpha)^{1/2} / 8 \equiv 10.1$. Here eq. (20) has been expressed in the original units using eq. (9), hence $U_{\max} = (4/3) \pi \sigma R^2$ in eq. (20).

We may conclude that the nucleation is governed by eq. (20) for $\omega \tau_1 > 1$ or

$$\omega > \tau_1^{-1} = 0.5 (\sigma / \rho \alpha)^{1/2} R_c^{3/2}. \quad (21)$$

Under this condition, appreciable droplet formation occurs for $(4 / \hbar \omega) U_{\max} \ln \Lambda \sim 80$, where $\Lambda = \omega \tau_1 / A_3 \alpha^{1/2}$ and $J_0$ is assumed to be on order of $e^{80}$, leading to

$$\omega \sim \left( \frac{1}{5} \ln \Lambda \right) \frac{1}{\hbar} \sigma R^2_c. \quad (22)$$

We may eliminate $R_c$ from eqs. (21) and (22) to obtain

$$\omega \gg \left( \frac{\sigma}{4 \rho \alpha} \right)^{2/7} \left( \frac{\sigma}{5 \hbar} \ln \Lambda \right)^{3/7}. \quad (23)$$

The right-hand side is 0.8$((1/5) \ln \Lambda)^{3/7} \times 10^{12}$ s$^{-1}$ for $^4$He and is 1.1$((1/5) \ln \Lambda)^{3/7} \times 10^{10}$ s$^{-1}$ for $^3$He-$^4$He (see A·10 for $\rho \alpha$). Therefore,
the experiment seems to be easier in $^3\text{He}-^4\text{He}$ than in $^4\text{He}$. Moreover, in $^3\text{He}-^4\text{He}$, $R_c$ can be made to be on the order of 10 Å by increasing the supersaturation in composition, whereas in $^4\text{He}$ only very large values of $R_c$ can be attained using realistic excess pressures.

Although we have treated helium, a similar theory can be constructed for anisotropic ferromagnets. In such systems counterparts of eqs. (18) and (20) also hold under oscillating magnetic field and $\tau_s$ can be long for weak anisotropy, so there seem to be a rich variety of new experiments. Details will be reported in the near future.

Acknowledgements

I would like to thank Professor T. Mizusaki for valuable discussions. A lecture by Professor S. Takagi was very informative in an early stage of the present work.

Appendix: Droplet Kinetics in $^4\text{He}$

Due to the incompressibility of the liquid region, the velocity field $v_l$ outside a spherical solid domain is given by

$$v_l = A \frac{1}{r} \nabla \frac{1}{r}, \quad (A \cdot 1)$$

where $A$ is determined from the mass conservation $w = \rho_l (v_l - \dot{R}) = \rho_s (v_s - \dot{R}) = -\rho_s \ddot{R}$ at the interface $r = R$ as

$$A = (1 - \rho_s / \rho_l) R^3 \dot{R}. \quad (A \cdot 2)$$

The velocity $v_s$ inside the droplet has been assumed to vanish. Then the momentum equation $\dot{v}_l + (v_l \cdot \nabla v_l) = -\rho_s^{-1} \nabla p$ in the liquid region becomes

$$p(r, t) - p_\infty = \rho_l \left[ \frac{1}{r} \dot{A} - \frac{1}{2r^4} \dot{A}^2 \right], \quad (A \cdot 3)$$

where $p_\infty$ is the pressure far from the droplet. As $r \rightarrow R$ the pressure $p_+$ immediately outside the droplet is related to $R$ by

$$p_+ = p_\infty - (\rho_s - \rho_l) \left[ R \ddot{R} + \left( \frac{3}{2} + \frac{\rho_s}{2\rho_l} \right) \dot{R}^2 \right]. \quad (A \cdot 4)$$

As the boundary conditions of the pressure $p$ and the chemical potential $\mu$, we require

$$[p + \rho (v - \dot{R})^2] = -\sigma \kappa, \quad (A \cdot 5)$$

$$\left[ \mu + \frac{1}{2} (v - \dot{R})^2 \right] = 0, \quad (A \cdot 6)$$

where $[\cdots]$ denotes the discontinuity across the interface and $\kappa = 2/R$ is the curvature. These relations readily follow from the balance of momentum and energy at $r = R$ in the local reference frame moving with the interface. Because $\delta p = \rho \delta \mu$ for the deviations at low temperatures, the pressures, $p_+$ and $p_-$, immediately outside and inside the interface are obtained as

$$p_+ = p_\infty + (\rho_s / (\rho_s - \rho_l)) \sigma \kappa - \frac{1}{2} \left( \frac{1}{\rho_l} - \frac{1}{\rho_s} \right) w^2, \quad (A \cdot 7)$$

$$p_- = p_\infty + (\rho_s / (\rho_s - \rho_l)) \sigma \kappa + \frac{1}{2} \left( \frac{1}{\rho_l} - \frac{1}{\rho_s} \right) w^2, \quad (A \cdot 8)$$

where $w$ is the mass current through the interface and is equal to $-\rho_s \dot{R}$ in the present case. Note that eqs. (A·7) and (A·8) are generally valid for arbitrary $\kappa$ and $w$ in the dissipation-free regime. Uwaha and Nozières already obtained the corresponding relations for the planar interface case. We substitute eq. (A·7) into eq. (A·4) to obtain the Rayleigh-Plesset equation,

$$\rho_{\text{eff}} \left( R \ddot{R} + \frac{3}{2} \dot{R}^2 \right) = -\frac{2\sigma}{R} + \mu_{\text{eff}}, \quad (A \cdot 9)$$

where $\rho_{\text{eff}}$ and $\mu_{\text{eff}}$ are given by eqs. (4) and (5). It is easy to verify that $H$ given by eq. (1) is a constant of motion when $\mu_{\text{eff}}$ is independent of time.

So far we have assumed that the velocity inside the droplet vanishes. However, when a gas droplet is considered, $w \equiv 0$ or all the velocities coincide at the interface. In this case, due to high compressibility of bubbles, both the droplet size and the acoustic field are strongly disturbed when the acoustic frequency approaches a particular resonance frequency. Similar resonance may also be expected against an ac electric field and such an experiment seems to be interesting.

Generalization of the above results to the mixture case is straightforward. In particular, the effective mass density $\rho_{\text{eff}}$ in $^3\text{He}-^4\text{He}$ is much larger than in $^4\text{He}$ because of the small-
ness of the $^3\text{He}$ concentration $c = \rho_3 / \rho$ in the $^4\text{He}$-rich metastable region. The $^3\text{He}$ velocity $v_3$ at the interface $r = R$ satisfies $\rho_3 (v_3 - \dot{R}) = -\rho_3 \dot{R}$ from the conservation law, where $\rho_3$ is the $^3\text{He}$ mass density of the $^3\text{He}$-rich phase. Here we note that $\rho_3 = c \rho - c \rho_3^*$, so that $|v_3|$ is larger than $|\dot{R}|$ by $1/c$. We assume the incompressibility of $^3\text{He}$ outside the droplet. Then $v_3$ decays as $R^2/r^2$ for $r > R$. Thus the kinetic energy of $^3\text{He}$ outside the droplet is calculated as

$$E_3 \approx 2\pi \rho_3^* (1 - \rho_3^* / \rho_3)^2 R^3 \dot{R}^2, \quad (A \cdot 10)$$

where $\rho_3^*$ is the effective mass density of the Fermi particles. Then $m_3^* = m_3 \rho_3^* / \rho_3$ is the effective mass of a Fermi particle. We expect that the ratio $\rho_3^* / \rho_3 = m_3^* / m_3$ is of order 1. The kinetic energy $E_4$ of $^4\text{He}$ is smaller than $E_3$ by the factor $c$ because $\rho_3 v_3 + \rho_4 v_4$ is small in our case. It follows that

$$\rho_{\text{eff}} = \rho_3^* (1 - \rho_3^* / \rho_3)^2 - \rho / c. \quad (A \cdot 11)$$

We find $\rho_{\text{eff}} \sim 2 \text{ g/cm}^3$.

References