

Quantum Nucleation in Helium under High-Frequency Field

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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 τ_s . In particular, we present a new formula for the quantum nucleation rate in the limit $\omega\tau_s \gg 1$.

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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 τ_s .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)}

$$H = 2\pi\rho_{\text{eff}}R^3\dot{R}^2 + 4\pi\sigma R^2 - \frac{4\pi}{3}\mu_{\text{eff}}R^3, \quad (1)$$

where ρ_{eff} is a mass density, $\dot{R} = \partial R / \partial t$, σ is the surface tension, and μ_{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 μ_{eff} is slightly positive, the liquid is metastable and the critical radius R_c is given by

$$R_c = 2\sigma / \mu_{\text{eff}}. \quad (2)$$

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

$$v_\infty = [2\mu_{\text{eff}} / 3\rho_{\text{eff}}]^{1/2}, \quad (3)$$

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

$$\rho_{\text{eff}} = (\rho_s - \rho_l)^2 / \rho_l \quad (4)$$

$$\mu_{\text{eff}} = (\rho_s / \rho_l - 1)(p - p_{cx}), \quad (5)$$

where ρ_s and ρ_l are the mass densities of the solid and the liquid, respectively, p is the pressure in the metastable liquid, and p_{cx} is the

pressure (~ 25 atm) of equilibrium two-phase coexistence.

When a sound wave is present, the pressure deviation $\delta p = p - p_{cx}$ consists of a constant (dc) part and an oscillating (ac) part as $\delta p = \bar{\delta p} + p_1 \cos(\omega_a t)$. We may also apply an ac electric field $E(t) = E_0 \cos(\omega_e t)$ to a metastable helium system. Because the dielectric constant ϵ_i within a droplet is slightly different from that (ϵ_0) in the surrounding medium, a dipolar field is induced around a droplet under $E(t)$.⁷ The disturbance changes on the spatial scale of R , so that the excess energy ΔH is proportional to $R^3 E(t)^2 = 1/2 R^3 E_0^2 [1 + \cos(2\omega_e t)]$. Because $\Delta H \propto R^3$, ΔH only changes μ_{eff} in eq. (1). Therefore, both in acoustic and electric fields, μ_{eff} may be expressed as

$$\mu_{\text{eff}} = \bar{\mu}_{\text{eff}} [1 + \epsilon \cos(\omega t)]. \quad (6)$$

To correctly calculate ΔH or the parameter ϵ 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,

$$x = (2R/3R_c)^{5/2}, \quad (7)$$

where R_c is the critical radius in eq. (2) for the dc chemical potential $\bar{\mu}_{\text{eff}}$. It should not be confused with the space coordinate. The time t will be scaled by $\tau_0 = (3/5)(3\rho_{\text{eff}}/2\sigma)^{1/2} R_c^{3/2}$, ω by τ_0^{-1} , and the energy by $E_0 = 9\pi\sigma R_c^2$. Then we obtain

$$H^* = H/E_0 = \frac{1}{2}x^2 + x^{4/5} - x^{6/5} [1 + \epsilon \cos(\omega t)]. \quad (8)$$

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

$$\begin{aligned} \hbar_{\text{eff}} &= \hbar/(\tau_0 E_0) \\ &= (5\hbar/8\pi)(\rho_{\text{eff}}\sigma)^{-1/2} \left(\frac{3}{2}R_c\right)^{-7/2}. \end{aligned} \quad (9)$$

For realistic values of R_c (~ 10 Å), \hbar_{eff} remains extremely small. Let us consider the ground-state wave function for the Hamiltonian $-(1/2)\hbar_{\text{eff}}^2(\partial/\partial x)^2 + x^{4/5}$ in which the chemi-

cal potential part is neglected. Its width is on order of $x_Q = \hbar_{\text{eff}}^{5/7}$ 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 $R_Q = x_Q^{2/5} R_c \sim (5\hbar/8\pi\sqrt{\rho_{\text{eff}}\sigma})^{2/7}$ (~ 1 Å) 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

$$i\hbar_{\text{eff}} \frac{\partial}{\partial t} \Psi = H^* \Psi, \quad (10)$$

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

$$\dot{S} + \frac{1}{2} S'^2 + U_0 + U_1 \cos(\omega t) = 0, \quad (11)$$

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

$$S_0 = i \int_0^x dx_1 [2U_0(x_1)]^{1/2}. \quad (12)$$

The deviation $S_1 = S - S_0$ obeys

$$\dot{S}_1 + i(2U_0)^{1/2} S_1' + \frac{1}{2} S_1'^2 + U_1 \cos(\omega t) = 0. \quad (13)$$

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 ,

$$\tau(x) = \int_0^x dx_1 [2U_0(x_1)]^{-1/2}. \quad (14)$$

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

$$\left(\frac{\partial}{\partial t} + i\frac{\partial}{\partial \tau}\right) S_1 + U_1 \cos(\omega t) \cong 0. \quad (15)$$

Because $\partial/\partial t + i\partial/\partial \tau = i(\partial/\partial \tau)_\xi$ with $\xi = t + i\tau$ held constant, we obtain

$$S_1 \equiv i \int_0^x dx_1 [2U_0(x_1)]^{-1/2} U_1(x_1) \cos [\omega t + i\omega(\tau - \tau_1)], \quad (16)$$

where x_1 is the integration variable and $\tau_1 = \tau(x_1)$. It is important that, for $\omega\tau \sim \omega x^{3/5} \gg 1$, or for $x \gg x_\omega = \omega^{-5/3}$, S_1 grows exponentially with increasing x as

$$S_1 \cong -i\epsilon A_1 \omega^{-3} \exp[-i\omega t + \omega\tau(x)], \quad (17)$$

where $A_1 = 18/25\tau_0^3$. We may confirm the following for $\omega\tau_s \geq 1$: (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 $(\partial/\partial t + i\partial/\partial\tau)S_1 \cong (-i\omega + i\partial/\partial\tau)S_1 \cong 0$ 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 $(1/2)S_1^2$ 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 (\propto the time average of $|\bar{\Psi}|^2$ at $x=1$) may be expressed for $\omega\tau_s \geq 1$ as

$$J = J_0 \exp \left[- \left(\frac{25\pi}{64\sqrt{2}\hbar_{\text{eff}}} \right) \{ 1 - A_2 \epsilon (\omega\tau_s)^{-3} e^{\omega\tau_s} \} \right], \quad (18)$$

where $J_0 \sim e^{80}$ is a microscopic constant and $A_2 = 9\pi^2/5 \cong 17.8$. We note that J coincides with the original expression by Lifshitz and Kagan for $\epsilon=0$ and that the oscillating perturbation becomes appreciable for $\omega\tau_s \gg 1$ due to the exponential factor $\exp(\omega\tau_s)$ however small ϵ is. Ivlev and Mel'nikov have generally discussed similar exponential enhancement of the effect of high-frequency electric field on the tunnelling rate.⁶⁾

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

$$\Psi = \sum_{n=0}^{\infty} \frac{1}{n!} [\epsilon A_1 / \omega^3 \hbar_{\text{eff}}]^n \exp \left[-in\omega t - \frac{1}{\hbar_{\text{eff}}} \int_0^x dx_1 \sqrt{2U_0(x_1) - 2\hbar_{\text{eff}}n\omega} \right]. \quad (19)$$

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_{eff} , S_0 and S_1 given by eqs. (12) and (17) may be reproduced. Equation (19) is a superposition of excited states with energy $E_n = n\hbar\omega$, and the low-energy states with $\hbar_{\text{eff}}n\omega \ll U_0(x)$ 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 $R_c/(\omega\tau_s)^{2/3}$ at which $\omega\tau(x) \sim 1$. Summing up the terms with $n > U_{\text{max}}/\hbar\omega$ in eq. (19), we find another nucleation rate from these highly excited states,

$$J' = J'_0 \exp \left[- \frac{4}{\hbar\omega} U_{\text{max}} \ln(\omega\tau_s/A_3\epsilon^{1/2}) \right], \quad (20)$$

where J'_0 is a microscopic quantity and $A_3 = 9\pi(3e)^{1/2}/8 \cong 10.1$. Here eq. (20) has been

expressed in the original units using eq. (9), hence $U_{\text{max}} = (4/3)\pi\sigma R_c^2$ in eq. (20).

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

$$\omega \gg \tau_s^{-1} = 0.5(\sigma/\rho_{\text{eff}})^{1/2} R_c^{-3/2}. \quad (21)$$

Under this condition, appreciable droplet formation occurs for $(4/\hbar\omega)U_{\text{max}} \ln \Lambda \sim 80$, where $\Lambda = \omega\tau_s/A_3\epsilon^{1/2}$ and J'_0 is assumed to be on order of e^{80} , leading to

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

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

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

The right-hand side is $0.8((1/5) \ln \Lambda)^{3/7} \times 10^{12} \text{ s}^{-1}$ for ${}^4\text{He}$ and is $1.1((1/5) \ln \Lambda)^{3/7} \times 10^{10} \text{ s}^{-1}$ for ${}^3\text{He}$ - ${}^4\text{He}$ (see (A·10) for ρ_{eff}). 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 \AA 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 τ_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.

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

Due to the incompressibility of the liquid region, the velocity field v_1 outside a spherical solid domain is given by

$$v_1 = A \frac{1}{r^3} r = -A \nabla \frac{1}{r}, \quad (\text{A} \cdot 1)$$

where A is determined from the mass conservation $w = \rho_1(v_1 - \dot{R}) = \rho_s(v_s - \dot{R}) = -\rho_s \dot{R}$ at the interface $r=R$ as

$$A = (1 - \rho_s/\rho_1) R^2 \dot{R}. \quad (\text{A} \cdot 2)$$

The velocity v_s inside the droplet has been assumed to vanish. Then the momentum equation $\dot{v}_1 + (v_1 \cdot \nabla) v_1 = -\rho_1^{-1} \nabla p$ in the liquid region becomes

$$p(r, t) - p_\infty = \rho_1 \left[\frac{1}{r} \dot{A} - \frac{1}{2r^4} A^2 \right], \quad (\text{A} \cdot 3)$$

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

$$p_+ = p_\infty - (\rho_s - \rho_1) \left[R \ddot{R} + \left(\frac{3}{2} + \frac{\rho_s}{2\rho_1} \right) \dot{R}^2 \right]. \quad (\text{A} \cdot 4)$$

As the boundary conditions of the pressure p and the chemical potential μ , we require

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

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

where $[\dots]$ 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

$$p_+ = p_{cx} + [\rho_1/(\rho_s - \rho_1)] \sigma \kappa - \frac{1}{2} \left(\frac{1}{\rho_1} - \frac{1}{\rho_s} \right) w^2, \quad (\text{A} \cdot 7)$$

$$p_- = p_{cx} + [\rho_s/(\rho_s - \rho_1)] \sigma \kappa + \frac{1}{2} \left(\frac{1}{\rho_1} - \frac{1}{\rho_s} \right) w^2, \quad (\text{A} \cdot 8)$$

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 κ and w in the dissipation-free regime. Uwha and Nozières already obtained the corresponding relations for the planar interface case.⁸⁾ We substitute eq. (A·7) into eq. (A·4) to obtain the Rayleigh-Plesset equation,

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

where ρ_{eff} and μ_{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 μ_{eff} is independent of time.

So far we have assumed that the velocity inside the droplet vanishes. However, when a gas droplet is considered, $w \cong 0$ 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.⁹⁾ 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 ρ_{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 ρ'_3 is the ${}^3\text{He}$ mass density of the ${}^3\text{He}$ -rich phase. Here we note that $\rho_3 = c\rho \sim c\rho'_3$, 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

$$E_3 \cong 2\pi\rho_3^*(1 - \rho'_3/\rho_3)^2 R^3 \dot{R}^2, \quad (\text{A} \cdot 10)$$

where ρ_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

$$\rho_{\text{eff}} \cong \rho_3^*(1 - \rho'_3/\rho_3)^2 \sim \rho/c. \quad (\text{A} \cdot 11)$$

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

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