Nucleation in dilute $^3$He-$^4$He liquid mixtures at low temperatures

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Abstract

We present a study of phase separation from supersaturated $^3$He-$^4$He liquid mixtures at low temperatures addressing both the degree of critical supersaturation $\Delta x_{cr}$ and the thermal-to-quantum crossover temperature $T^\ast$ for the nucleation process. Two different nucleation seeds are investigated, namely $^3$He droplets and $^4$He vortex lines with cores filled with $^3$He. We have found that the experimental $T^\ast$ is reproduced when we consider that nucleation proceeds from $^3$He droplets, whereas $\Delta x_{cr}$ is reproduced when we consider $^4$He vortex lines filled with $^3$He. However, neither nucleation configuration is able to simultaneously reproduce the current experimental information on $\Delta x_{cr}$ and $T^\ast$. 
I. INTRODUCTION

Supersaturated $^3$He-$^4$He liquid mixtures have been recognized as an ideal system to study nucleation phenomena. On the one hand, these mixtures can be made extremely pure, and on the other hand, $^4$He covers the walls of the experimental cells, preventing undesired heterogeneous nucleation that is difficult to avoid in other substances. Moreover, the fact that helium remains liquid up to zero temperature ($T=0$) has raised expectations that the transition from thermal to quantum nucleation could be observed as $T$ decreases.

The first experiments on metastable helium mixtures were carried out in the sixties, but one had to wait more than twenty years for a systematic study of this phenomenon. Meanwhile, the kinetics of nucleation in dilute helium mixtures was theoretically addressed by Lifshitz et al., much along the line of the classical work of Lifshitz and Kagan.

At very low temperature and at zero pressure, the concentration of $^3$He in the mixture at saturation is $x_s \sim 6.6\%$. However, supersaturated $^3$He-$^4$He mixtures can be found in a metastable state for concentrations $x$ above the saturation value. A first estimate of the degree of supersaturation $\Delta x_{cr} \equiv x - x_s$ resulted from extrapolation of the measured $^3$He chemical potential excess $\Delta \mu_3$ along the demixing line. This extrapolation yielded $\partial \Delta \mu_3 / \partial x \geq 0$ up to values of the $^3$He concentration $x \gtrsim 16\%$, thus giving $\Delta x_{cr} \sim 10\%$. Large values of critical supersaturation have also been obtained in microscopic and density functional calculations.

An intriguing observation is the small degree of critical supersaturation attained in recent experiments on phase separation in supersaturated helium mixtures. It was found to be of the order of 1% in the experiments carried out by the Ukrainian group, and below 0.5% in those of the Japanese group. As a matter of fact, the measurements in the sixties were also yielding small $\Delta x_{cr}$.

A possible explanation of the discrepancy between theory and experiment is that phase separation at small supersaturations may be triggered by heterogeneous nucleation on vortex lines. Indeed, at the low temperatures involved in the experiments $^4$He is still superfluid due to the limited solubility of $^3$He into $^4$He. This mechanism was proposed in Ref. [15], and it has been further elaborated. It turns out that the presence of vortex lines in the mixture decreases the degree of critical supersaturation from a value $\Delta x_{cr} \sim 10\%$ to $\sim 1\%$. At present, both experimental groups seem to have adhere to this explanation of their results.

Besides critical supersaturation, another experimental quantity of most interest is the temperature at which the nucleation process changes from being a thermal one to a quantal one. This crossover temperature $T^*$ has been determined by measuring the temperature below which the degree of critical supersaturation becomes almost $T$ independent; it is of the order of 10 mK. Obviously, the theoretical description of the process cannot be considered as achieved until $\Delta x_{cr}$ and $T^*$ are satisfactorily obtained within the same framework.

The present aim is to complete our previous works with the calculation of the crossover temperature using the same basic ingredients that we have used to obtain the critical supersaturation. To this end, we have resorted to a dynamical method, namely the functional-integral approach (FIA), that proved to be well suited to quantitatively describe quantum cavitation in liquid $^4$He.
II. FORMALISM

The segregation of $^3$He from a dilute $^3$He-$^4$He liquid mixture at low $T$ is a first order phase transition. This means that in order to nucleate the pure $^3$He phase, the system has to overcome an energy barrier $\Delta \Omega$. Thus, the mixture may be at $^3$He concentrations above the one corresponding to the two-phase equilibrium value at the given pressure ($P$) and temperature $T$.

At high enough $T$, segregation is thermally activated and the energy barrier is overcome by the energy provided to the system by a thermal bath. In this case, the nucleation rate, i.e., the number of critical nucleation seeds formed per unit volume and time is given by

$$J_T = J_0 T \exp\left[\frac{-\Delta \Omega_{\text{max}}}{k_B T}\right],$$

(1)

where the prefactor $J_0 T$ depends on the dynamics of the nucleation process, $k_B$ is the Boltzmann’s constant and $\Delta \Omega_{\text{max}}$ is the height of the energy barrier. At low $T$, segregation by thermal activation is no longer possible. However, it can proceed by quantum tunneling: the metastable liquid mixture ‘tunnels’ through the energy barrier. In the limit of zero temperature, the transition is purely quantal, but below $T^*$ thermally assisted tunneling also occurs. This is one of the many thermally assisted quantum tunneling processes occurring in physics (see for example Ref. [23] and refs. therein). For $T < T^*$ the tunneling rate is

$$J_Q = J_{0Q} \exp(-S^Q),$$

(2)

where $\exp(-S^Q)$ is the tunneling probability, and the prefactor $J_{0Q}$ is of the order of the number of nucleation sites per unit volume times an attempting frequency.

To obtain the tunneling probability one formally starts from the statistical average of the transition probability over a time $t = t_f - t_i$:

$$P = \sum_{i,f} \frac{|\langle \Psi_f | \exp\left[-\frac{i}{\hbar} \int_{t_i}^{t_f} dt \hat{H} \right] | \Psi_i \rangle|^2 \exp(-E_i/k_B T)}{\sum_i \exp(-E_i/k_B T)},$$

(3)

where $\hat{H}$ is the Hamiltonian of the system, $E_i$ are the energy states, and $\Psi_i$ and $\Psi_f$ are the wave functions of the initial and final states. This expression can be written in a more workable form by using the path integral formulation of quantum mechanics, and its connection with statistical mechanics. According to Feynman [24] (see also Refs. [25–27]), $P$ can be written as the functional integral

$$\int D[q(\tau)] \exp\left[-\frac{1}{\hbar} \oint d\tau \mathcal{L}[q(\tau)]\right],$$

(4)

where $\mathcal{L}[q(\tau)]$ is the imaginary-time ($\tau = it$) classical Lagrangian of the system and $D[q(\tau)]$ denotes integration over all periodic trajectories $q(\tau)$ with period $\tau_p = \hbar/k_B T$ in the potential well that results from inverting the energy barrier. The integral in the exponent of Eq. (4) is the imaginary-time action $S(T) = \oint d\tau \mathcal{L}$ evaluated over the period $\tau_p$. In the semiclassical limit $S(T) >> \hbar$, the trajectory that contributes the most from all possible periodic orbits is the one which minimizes the action. It leads to Eq. (2) with $S^Q = S_{\text{min}}(T)/\hbar$, where $S_{\text{min}}$ is the minimum action.
The practical usefulness of Eq. (4) for the problem at hand is that physical insight allows one to guess $L$ as a functional of densities and collective velocities instead of dealing with the impracticable Eq. (3). To implement this scheme, it is quite clear that a sound approximation is needed for the imaginary-time action. This means a realistic energy barrier and a simple yet reliable choice of the integration path $q(\tau)$.

Let us now work out in some detail the case in which only one collective coordinate $\delta$ is considered. It is then a simple task to minimize the imaginary-time effective action:

$$S(T) = \oint d\tau L[\delta(\tau)] = \int_{-\tau_p/2}^{\tau_p/2} d\tau \left[ \frac{1}{2} M(\delta) \dot{\delta}^2 + \Delta\Omega(\delta) \right]. \tag{5}$$

As indicated, the effect of continuing the action to imaginary time is to invert the ‘potential’, i.e., $\Delta\Omega \rightarrow -\Delta\Omega$ in the equation of motion, and the identification of $k_B T$ with $\hbar/\tau_p$; the path $\delta(\tau)$ defined in imaginary time $\tau$ has to fulfill the periodic boundary condition $\delta(-\tau_p/2) = \delta(\tau_p/2)$. This is illustrated in the schematic Figure 1. We have supposed that the collective mass $M$ depends on $\delta$, which will be the practical case. Imposing the extremum condition on the action yields the following equation of motion for $\delta(\tau)$:

$$M(\delta)\ddot{\delta} + \frac{1}{2} \frac{dM}{d\delta} \dot{\delta}^2 = \frac{d\Delta\Omega}{d\delta}. \tag{6}$$

Multiplying Eq. (6) by $\dot{\delta}$ we have

$$\frac{d}{d\tau} \left[ \frac{1}{2} M(\delta) \dot{\delta}^2 - \Delta\Omega(\delta) \right] = 0. \tag{7}$$

Thus

$$\frac{1}{2} M(\delta) \dot{\delta}^2 - \Delta\Omega(\delta) = \text{constant} \equiv -E \tag{8}$$

with $\Delta\Omega_{\text{max}} \geq E \geq 0$.

Eq. (8) has the trivial solution $\delta = \delta_0$ corresponding to the minimum of $-\Delta\Omega$: the system is ‘at rest’ at the bottom of the inverted barrier potential well. In this case, $E = \Delta\Omega_{\text{max}}$, and the integration of Eq. (8) yields

$$S^Q = \frac{1}{\hbar} S_{\text{min}}(T) = \frac{\Delta\Omega_{\text{max}}}{\hbar} \int_{-\tau_p/2}^{\tau_p/2} d\tau = \frac{\Delta\Omega_{\text{max}}}{k_B T}. \tag{9}$$

Thus, the trivial solution yields the exponent for classical thermal activation, see Eq. (1). It means that within FIA, the transition between thermal and quantal regime is smooth. For $E < \Delta\Omega_{\text{max}}$, one has to seek periodic solutions $\delta(\tau)$ whose turning points $\delta_1$ and $\delta_2$ are such that $\Delta\Omega(\delta_1) = \Delta\Omega(\delta_2) = E$ (see Fig. 1). Integrating Eq. (8) we get the period $\tau_p$:

$$\tau_p(E) = 2 \int_{\delta_1(E)}^{\delta_2(E)} d\delta \sqrt{\frac{M(\delta)}{2[\Delta\Omega(\delta) - E]}}. \tag{10}$$

Using
\[
d\tau = \sqrt{\frac{M(\delta)}{2[\Delta\Omega(\delta) - E]}} \, d\delta \tag{11}
\]

the action Eq. (3) becomes

\[
S^Q(E) = \frac{2}{\hbar} \int_{\delta_1(E)}^{\delta_2(E)} d\delta \left[ 2\Delta\Omega(\delta) - E \right] \sqrt{\frac{M(\delta)}{2[\Delta\Omega(\delta) - E]}}. \tag{12}
\]

At \( T = 0 \), \( \tau_p = \infty \) and \( E = 0 \). In this case, the solution to Eq. (8) is the usual instanton\(^2\), and \( S^Q(E = 0) \) coincides with the WKB approximation at zero energy\(^3\):

\[
S^{WKB} = \frac{2}{\hbar} \int_{\delta_1(E=0)}^{\delta_2(E=0)} d\delta \sqrt{2M(\delta)\Delta\Omega(\delta)}. \tag{13}
\]

The crossover temperature is obtained by equating Eqs. (1) and (2) we get:

\[
S^Q(E = \Delta\Omega_{\text{max}}) = \frac{\Delta\Omega_{\text{max}}}{k_B T^*}. \tag{14}
\]

Taking a trajectory \( \delta \) corresponding to \( E \approx \Delta\Omega_{\text{max}} \) and using Eq. (12), we can write

\[
S^Q(E \approx \Delta\Omega_{\text{max}}) \approx \frac{2}{\hbar} \Delta\Omega_{\text{max}} \int_{\delta_1(E \approx \Delta\Omega_{\text{max}})}^{\delta_2(E \approx \Delta\Omega_{\text{max}})} d\delta \left[ M(\delta) \right] \sqrt{\frac{2}{[\Delta\Omega(\delta) - E]}}. \tag{15}
\]

The comparison to Eq. (10) yields \( \hbar / (k_B T^*) = \tau_p(E \approx \Delta\Omega_{\text{max}}) \). An analytical expression for \( T^* \) is obtained by expanding Eq. (14) around the maximum of \( \Delta\Omega \) located at \( \delta_0 \). It reads

\[
k_B T^* = \frac{\hbar}{2\pi} \left[ \frac{1}{M(\delta_0)} \frac{d^2 \Delta\Omega}{d\delta^2} \right]_{\delta_0}. \tag{16}
\]

Eq. (16) shows that the value of \( T^* \) is determined by small variations around \( \delta_0 \). This is a well-known result that was suggested long ago by Goldanskii\(^2\), the validity of which goes beyond the simple model we have used to derive it. Generally speaking, the crossover temperature is determined from the frequency of the small amplitude oscillations around the minimum of the inverted barrier potential well. When more realistic methods are employed to generate the critical nucleation configurations, like for example density functional theory\(^3\),\(^4\), the problem becomes an infinite-dimensional one, whose solution is quite a formidable task. Crossover temperatures for thermally assisted quantum cavitation in liquid helium have been determined only recently\(^2\),\(^3\).

Finally, we mention that \( S^{WKB} \) can be used to estimate \( T^* \) through the expression\(^3\),\(^4\)

\[
k_B T^*_{WKB} = \frac{\Delta\Omega_{\text{max}}}{S^{WKB}}. \tag{17}
\]
III. ENERGY BARRIERS AND IMAGINARY-TIME ACTIONS

As indicated at the introduction, we have considered two different nucleation seeds, depending on whether there is some vorticity in the experimental sample or not. The first one is a pure $^3$He droplet embedded into the $^3$He-$^4$He mixture, and the second one is a $^4$He vortex line filled with pure $^3$He. In the following we work out in detail the energy barriers and imaginary time actions making use of the capillarity approximation in the case of a droplet, and of the hollow core approximation in the case of a vortex line. These are sound approximations provided nucleation occurs near the two phase equilibrium curve. Indeed, it is well known that near this curve, the critical nucleation configuration, i.e. that corresponding to the maximum of the energy barrier, is large. This makes negligible ‘finite size effects’ such as interface diffuseness and curvature corrections. This has been checked by comparing the capillarity model results with density functional calculations, which selfconsistently incorporate finite size effects\textsuperscript{14–16}. We recall that experimentally, the process occurs near the saturation line; otherwise, $\Delta x_{cr}$ would be large. This is at variance with the process of cavitation in liquid helium, which takes place near the spinodal line\textsuperscript{34–36}, where the cavitation seeds are small, pachydermic bubbles and the capillarity approximation fails\textsuperscript{21,30,37}. In the case of nucleation triggered by $^3$He droplets, we will also present the results obtained using a density functional FIA as described in Ref. 32.

A. $^3$He droplets

Within the capillarity approximation, the energy barrier of a $^3$He droplet of radius $R$ immersed in the mixture is written as

$$\Delta \Omega(R) = 4\pi R^2 \sigma - \frac{4\pi}{3} R^3 \rho_{30} \Delta \mu_3 ,$$

(18)

where $\sigma$ is the surface tension of the $^3$He-$^4$He interface, $\rho_{30}$ is the density of pure $^3$He inside the droplet, and $\Delta \mu_3$ is the difference between the chemical potential of $^3$He in the metastable mixture and in pure $^3$He. This difference is negative when the mixture is stable and positive otherwise. Upon minimization with respect to $R$, the maximum of the energy barrier and the radius of the critical droplet $R_c$ are given by

$$\Delta \Omega_{\text{max}} = \frac{4\pi}{3} \sigma R_c^2 = \frac{16\pi}{3} \frac{\sigma^3}{(\rho_{30} \Delta \mu_3)^2}$$

$$R_c = 2\sigma/(\rho_{30} \Delta \mu_3) .$$

(19)

Values of $R_c$ and $\Delta \Omega_{\text{max}}$ can be obtained using the experimental values\textsuperscript{2,38–39} of the physical quantities entering their definition. In particular, the experimental data\textsuperscript{2} can be employed to write the above equations as a function of $\Delta x$ using the approximation $\Delta \mu_3 \approx (\partial \Delta \mu_3 / \partial x) \Delta x$ along the demixing line. For instance, at zero pressure $\Delta \mu_3 \sim 2.49 \Delta x$ K. In view of the weak $T$ dependence of these quantities in the range of temperatures of experimental interest (below 150 mK), it is legitimate to consider their values at zero K. If nucleation proceeds thermally, the degree of critical supersaturation at fixed $P$ and $T$ is obtained by solving the equation $1 = (V \cdot t)_{\text{exp}} J_T$. It determines $\Delta x_{cr}$ from $\Delta \Omega_{\text{max}}$ (Eq. (19)):
\[ \Delta \Omega_{\text{max}} = k_B T \ln [(V \cdot t)_{\text{exp}} J_{0T}] . \]  

(20)

We have used as prefactor an attempting frequency per unit volume \( J_{0T} = \nu_0/V_0 \sim (k_B T/h)/(4\pi R_0^3/3) \). As pointed out, the prefactor depends on the physics of the nucleation process, but the results are not very sensitive to the precise value of the product \((V \cdot t)_{\text{exp}} J_{0T}\) (see the discussion after Eq. (20)). Yet, using the estimates made at the end of Ref. 32, we have checked that in the present case, where phase separation proceeds by the diffusion of \(^3\)He atoms, the diffusion time to make a critical \(^3\)He droplet of \(R_0 \sim 10\AA \) is, in the worst case, of the same order of magnitude than the ‘thermal period’ \( \tau_T = 2\pi h/k_BT \). As thoroughly discussed by Burmistrov et al., care should be exerted when describing later stages in the dynamical evolution of the critical droplet after it reaches a mesoscopic size. Taking for the experimental volume and time \((V \cdot t)_{\text{exp}} \) those of Ref. 35 and for \(R_0 \sim 10\AA \), we obtain \( \Delta x_{cr} \sim 13\% \) at \( T = 100 \) mK, and \( \sim 19\% \) at \( T = 50 \) mK. This result is an order of magnitude larger than the experimental data. This is a drawback of using \(^3\)He droplets as nucleations seeds detected immediately after the first systematic experiments.

Let us now calculate the crossover temperature. In the following, we write \( R = R_c + \delta \), where here \( \delta \) measures the displacement of the surface of the drop from the value corresponding to the critical configuration. In this way, the dynamics of the nucleation process is described by the time evolution of \( \delta \). The energy barrier then reads

\[ \Delta \Omega(\delta) = 4\pi(R_c + \delta)^2 \sigma - \frac{4\pi}{3}(R_c + \delta)^3 \rho_{30} \Delta \mu_3 . \]  

(21)

The next step is to express the imaginary-time Lagrangian in terms of the displacement \( \delta \) and its time derivative \( \dot{\delta} \). Since the ‘potential part’ is just \(-\Delta \Omega(\delta)\), we have only to derive the kinetic energy term. This last is given by

\[ E_{\text{kin}} = \frac{m_3}{2} \int d\vec{r} \rho_3(\vec{r}, t) \dot{\vec{u}}_3^2(\vec{r}, t) + \frac{m_4}{2} \int d\vec{r} \rho_4(\vec{r}, t) \dot{\vec{u}}_4^2(\vec{r}, t) \equiv \frac{1}{2} M(\delta) \dot{\delta}^2 , \]  

(22)

where \( \rho_q(\vec{r}, t) \) and \( \dot{\vec{u}}_q(\vec{r}, t) \) are respectively, the particle density and the collective velocity of isotope \( q \). The collective mass \( M(\delta) \) is determined as follows. For a given configuration characterized by a \( \delta \) value, the densities of the system are written as:

\[ \rho_4(r) \equiv \rho_{4c}(r) = \rho_{4m}[1 - \Theta(R_c + \delta - r)] = \rho_{4c}(r - \delta) \]

\[ \rho_3(r) \equiv \rho_{3c}(r) = \rho_{3m}[1 - \Theta(R_c + \delta - r)] + \rho_{30}\Theta(R_c + \delta - r) = \rho_{3c}(r - \delta) , \]  

(23)

where \( \rho_{qm} \) with \( q = 3, 4 \) is the particle density of each isotope in the metastable phase \( (x = \rho_{3m}/(\rho_{3m} + \rho_{4m})) \), and the subscript \( c \) refers to the critical configuration. This is schematically illustrated in Fig. 3. The dynamics comes into Eqs. (23) through the time dependence of \( \delta \), i.e., \( \rho_q(r, t) = \rho_{qc}(r - \delta(t)) \). Thus,

\[ \rho_q(\vec{r}, t) = -\dot{\delta} \frac{d}{dr} \rho_{qc}(r) . \]  

(24)

Assuming spherical symmetry during the process of growing, the densities and collective velocities only depend on the modulus of \( \vec{r} \), and it is possible to write \( u_q(r, t) \) as a function of \( \delta \) and \( \dot{\delta} \) formally integrating the continuity equation.
\[ \frac{\partial \rho_q}{\partial t} + \nabla (\rho_q \vec{u}_q) = 0 . \] (25)

This yields

\[ u_q(r, t) = -\frac{1}{r^2 \rho_q(r, t)} \int_0^r s^2 \dot{\rho}_q(s, t) ds . \] (26)

Using Eqs. (23) and (26) the mass parameter becomes

\[ M(\delta) = 4\pi \left[m_4 \rho_{4m} + m_3 \frac{(\rho_{30} - \rho_{3m})^2}{\rho_{3m}} \right] (R_c + \delta)^3 \]
\[ \equiv 4\pi M(R_c + \delta)^3 . \] (27)

The same expression has been derived in Refs. 18,33 using a different method.

By inserting the expressions for \( \Delta \Omega(\delta) \) and \( M(\delta) \) in Eq. (16), we straightforwardly obtain the crossover temperature

\[ k_B T^* = \frac{\hbar}{2\pi} \sqrt{\frac{1}{M R_c^3}} . \] (28)

Furthermore, Eq. (17) yields

\[ T_{WKB}^* = \sqrt{\frac{2}{3} \frac{512}{405} T^* \approx 1.03 T^*} . \] (29)

From the \( \Delta x_{cr} \) values quoted below Eq. (24), we estimate \( T^* \sim 16 \text{ mK} \) for \( \Delta x_{cr} = 13 \% \), and \( T^* \sim 28 \text{ mK} \) for \( \Delta x_{cr} = 19 \% \). These crossover temperatures are compatible with those experimentally found.\(^5\) We want to stress that these estimates are quantitative provided \( \Delta x \) is small, i.e., far enough from the spinodal line in the \( P-x \) plane, which is determined by the condition\(^4\) \( (\partial \mu_3/\partial x)_{P,T} = 0 \). Indeed, as the nucleation barrier approaches to zero at the spinodal line, so does \( T^* \), and the law \( T^* \sim (\Delta x)^{3/2} \) following from Eqs. (19) and (28) eventually breaks down. A similar breakdown occurs in the cavitation process.\(^30\),\(^41\) Fig. 3 displays \( T^*(\Delta x) \) obtained from equation (28) at \( P = 0 \) and 1 bar (dashed lines). The values of \( \rho_{3m} \) and \( \rho_{4m} \) entering Eq. (27) when \( x \) is much larger than \( x_s \) have been calculated by means of the density functional of Ref. \(^42\). Had we used the values at the saturation line, the resulting \( T^* \) would have been a factor of two smaller for \( \Delta x \) larger than \( \sim 5 \% \).

We have also obtained \( T^*(\Delta x) \) using density functional theory to describe the critical nucleation clusters, and the FIA for the nucleation dynamics.\(^32\) The results are displayed in Fig. 3 (solid lines). For completeness, and to clarify some aspects underlying our way of obtaining the collective mass in the capillarity approximation, we present in the Appendix the equations of motion used to determine \( T^* \).

It can be concluded from Fig. 3 that the \( T^*(\Delta x) \) function obtained using a density functional that accurately describes the thermodynamical properties of the liquid mixture\(^42\) has a maximum of \( \sim 25 \text{ mK} \) at \( \Delta x \sim 18 \% \). Consequently, at \( P = 0 \) above \( T \sim 25 \text{ mK} \) the nucleation of the \(^3\)He-rich phase always proceeds thermally. At variance with the capillarity method, the density functional approach is able to realistically describe critical nucleation configurations from the saturation to the spinodal point. It is worth to notice that the
crossover temperatures obtained using the density functional plus FIA tend to zero at the saturation and spinodal points, whereas those obtained from the capillarity approximation vanish only at saturation. The calculated spinodal $\Delta x$ value at $P \sim 0$ is $\sim 25\%$ (see Ref. 14 and also Fig. 3). The crudeness of the capillarity approximation when applied too far from the coexistence line can be assessed comparing the density profiles of the critical nucleation configuration of the density functional method versus the capillarity approximation. Fig. 4 shows them at $P = 0$ for $\Delta x_{cr} \sim 16\%$.

Figure 3 simply tells us that if the mixture is brought to a supersaturation degree $\Delta x$, nucleation will proceed by quantum tunneling below $T^*(\Delta x)$. To determine which of the $T^*(\Delta x)$ would correspond to a given experimental situation, one has to determine the critical degree of supersaturation $\Delta x_{cr}$, which corresponds to the largest one the mixture can sustain before $^3$He droplets nucleate at an appreciable rate. This is obtained by solving the equation

$$1 = (V \cdot t)_{exp} J$$

(30)

taking either $J = J_T$ for $T \geq T^*$ or $J = J_Q$ for $T \leq T^*$, and using as a prefactor $J_{00}$ the estimate $k_B T^*/(hV_0)$, where $V_0 = 4\pi R_0^3/3$ with $R_0 = 10\,\text{Å}$. Within the more accurate density functional FIA and using as experimental $(V \cdot t)_{exp}$ two extreme values, namely $10^{14}\,\text{Å}^3$ sec and $10^4\,\text{Å}^3$ sec, at $P = 0$ one gets $\Delta x_{cr} \sim 18\%$ and $T^* \sim 25\,\text{mK}$, and $\Delta x_{cr} \sim 16\%$ and $T^* \sim 23\,\text{mK}$ respectively, and very similar values at $P = 1\,\text{bar}$. The maximum of the thermal nucleation barriers $\Delta \Omega_{max}$ are $\sim 1\,\text{K}$ and $\sim 0.5\,\text{K}$, respectively.

B. $^4$He vortex lines filled with $^3$He

The possibility of considering vortex lines as seeds of a kind of heterogeneous nucleation in the mixture stems from the fact that the experimental sample, if treated in conventional fashion, is likely permeated ab initio by quantized vortices stabilized by surface pinning. These vortex lines, whose core is filled with $^3$He, are stable for $x \leq x_s$, and metastable otherwise. Within the hollow core model (HCM), is it very simple to obtain their structure. The energy per unit length of vortex line is

$$\Omega(R) = 2\pi R \sigma - \pi R^2 \rho_{30} \Delta \mu_3 + \pi n^2 \frac{\hbar^2}{m_4} \rho_{4m} \ln \left( \frac{R_\infty}{R} \right).$$

(31)

In this equation, $R$ is the radius of the vortex core, $R_\infty$ is a large enough radius at which velocity vanishes, $n = 1, 2, \ldots$ is the quantum circulation number, and the remaining variables have the same meaning as before. Minimization with respect to $R$ yields at most two extrema corresponding to the values

$$R = 2R_0 \frac{\mu_c}{\Delta \mu_3} \left[ 1 \pm \sqrt{1 - \frac{\Delta \mu_3}{\mu_c}} \right],$$

(32)

where

$$R_0 = \frac{n^2 \hbar^2 \rho_{30}}{2 \sigma m_4},$$

$$\mu_c = \frac{\sigma^2 m_4}{2 n^2 \hbar^2 \rho_{30} \rho_{4m}}.$$
\( R_0 = \lim_{\Delta \mu_3 \to 0} R \) is the radius of the vortex core at the demixing line, i.e., \( \Delta \mu_3 = 0 \). If the vortex is in the stable region of the \( P - x \) plane, \( \Delta \mu_3 \) is negative, only the minus sign in Eq. (32) makes sense and the corresponding \( R \) defines the radius of the stable vortex. If \( \Delta \mu_3 > 0 \), Eq. (32) yields two core radii. The smallest one \( R_\ast \) corresponds to a minimum of \( \Omega(R) \), and the largest one \( R_\ast \) to a maximum. This is, of course, the situation for given \( (P, T, \Delta x) \) values. Consequently, \textit{ab initio} stable vortex lines become metastable as the mixture is driven into the supersaturated region. These metastable vortex lines can trigger phase separation by thermal fluctuations if an energy per unit length equal to \( \Delta \Omega_{\text{max}} = [\Omega(R_\ast) - \Omega(R_\ast)] \) is subministrated to them. As before, calling \( R_c \) the radius \( R_\ast \) of the critical configuration and defining \( R = R_c + \delta \), the energy barrier per unit length reads

\[
\Delta \Omega(\delta) = 2\pi(R_c + \delta)\sigma - \pi(R_c + \delta)^2 \rho_{30} \Delta \mu_3 + \pi n^2 \frac{\hbar^2}{m_4} \rho_{4m} \ln \left( \frac{R_\infty}{R_c + \delta} \right) - \Omega(R_\ast),
\]

(34)

where \( \Omega(R_\ast) \) is a constant for fixed \( (P, T, \Delta x) \).

It is interesting to notice from Eq. (32) that if \( \Delta \mu_3 > \mu_c \) the vortex line loses its metastable character. Indeed, when \( \mu_3 = \mu_c \) one has \( R_\ast = R_\ast \): the energy barrier disappears and the vortex core freely expands triggering phase separation. At \( P = 0 \), taking \( n = 1 \), \( \rho_{4m} \sim 0.020 \text{Å}^{-3} \) and the experimental values for the other variables, one gets \( R_0 \sim 7.6 \text{Å} \) and \( \mu_c \sim 0.032 \text{K} \) which yields \( \Delta x_{cr} \sim 1.3\% \). It is remarkable that this good agreement with experimental supersaturation values comes from an extremely simple model. The smallness of the obtained \( \Delta x_{cr} \) fully justifies the use of the hollow core model to calculate \( \Delta x_{cr} \) and \( T^* \).

To determine \( T^* \) we proceed as in the preceeding subsection. Equation (22) gives now the kinetic energy per unit length, and the expressions (23) hold with \( r \) being the cylindrical radial coordinate. Again, the continuity equation in cylindrical coordinates can be formally integrated yielding

\[
u_q(r, t) = -\frac{1}{r \rho_q(r, t)} \int_0^r s \dot{\rho}_q(s, t) ds.
\]

(35)

The collective mass for this geometry is readily obtained:

\[
M(\delta) = 2\pi M(R_c + \delta)^2 \ln \left( \frac{R_\infty}{R_c + \delta} \right),
\]

(36)

where \( M \) is defined in Eq. (27). In deriving the vibrational collective mass we have not taken into account any contribution from the circulation of the superfluid component, as only small radial displacements have to be considered if one supposes that phase separation is triggered by the expansion of the vortex core. Finally, we have for the crossover temperature

\[
k_B T^* = \frac{h}{2\pi} \sqrt{\frac{1}{M} \frac{\Delta \mu_3 \rho_{30} - R_0 \sigma / R_c^2}{R_c^2 \ln \left( \frac{R_\infty}{R_c} \right)}}.
\]

(37)

Taking \( \ln(R_\infty/R_c) \sim 1 \), one gets \( T^* \sim 1 \text{mK} \) for \( \Delta x \sim 1\% \) at \( P = 0 \).
IV. SUMMARY

In this work we have calculated the crossover temperature and critical supersaturation of $^3\text{He}^-^4\text{He}$ mixtures at very low temperatures using a functional-integral method in conjunction with either a density functional or a capillarity approximation description of nucleation configurations. We have found that the existence of vorticity in the mixture would explain the small $\Delta x_{cr}$ measured in experiments, but fails to reproduce $T^*$, yielding values an order of magnitude smaller than experiment. Conversely, conventional nucleation seeds consisting in $^3\text{He}$ droplets do reproduce the experimental crossover temperatures but yield $\Delta x_{cr}$ values an order of magnitude larger than experiment.

In spite of its failure, we feel that a detailed exposure of the merits and drawbacks of the present method might help find alternative roads to address this longstanding problem and identify any missing physical ingredient relevant to its solution. Besides, we think that our application of the FIA to this problem is especially simple and transparent, which adds a pedagogical value to it.

A line of possible improvement might consists in introducing from the start a dynamics based on a zero rather than a first sound hydrodynamic description of the harmonic vibrations leading to $T^*$. The density functional we have used reproduces the Landau parameters of the mixture that are relevant for such a zero sound description in the long wavelength limit; among other quantities, $^3\text{He}$ effective mass and the incompressibilities of both isotopes have been taken into account as fitting quantities to fix the density functional parameters. Moreover, the nucleation barrier is known to be well described by the new generation of static density functional calculations presented in Refs. [21,30,32]. It would be quite natural to introduce the zero sound dynamics much along the fruitful procedure followed in nuclear physics to describe nuclear collective excitations with density-dependent nucleon-nucleon effective interactions [44]. Such a program, which involves solving the Random-Phase-Approximation equations in imaginary time, has been formally developed to study a problem having some similarities to ours, namely spontaneous nuclear fission below the fission barrier [45]. Here and there, a practical implementation of this scheme is quite a formidable task, especially in presence of vortices.

Finally, we would like to point out that the experience gathered in the description of the nuclear giant monopole resonance indicates that for this precise mode, first and zero sound dynamics yield quite equivalent results, see chapter 13 of Ref. [44] and Ref. [46]. Thus, in the case of nucleation driven by $^3\text{He}$ droplets, as far as the problem of determining $T^*$ is amenable to study spherically symmetric, radial vibrations of the system, we expect that both dynamics yield rather similar $T^*$.

V. ACKNOWLEDGMENTS

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APPENDIX:

In this Appendix we give some hints about how we have derived the dynamical equations leading to $T^*$ in the case of $^3\text{He}$-$^4\text{He}$ mixtures. We recall that by construction, the static density functional assumes that $^3\text{He}$ is in the normal phase, and all $^4\text{He}$ is superfluid. For this reason, the 'two fluid' hydrodynamics of superfluids, as described for instance in Ref. 17, is of no applicability here.

The first step is to write the static density functional of the kind proposed by Dalfovo (see also Refs 14, 42) in a Galilean invariant form:

$$\mathcal{E}(\rho_q, \tau_q, \vec{j}_q) = \frac{1}{2m_3} \tau_3 + \frac{1}{2m_4} \tau_4 + g_3(\rho_3, \rho_4)(\rho_3 \tau_3 - \vec{j}_3^2) + g_4(\rho_3, \rho_4)(\rho_4 \tau_4 - \vec{j}_4^2)$$

$$+ g_{34}(\rho_3, \rho_4)(m_3^2 \rho_3 \tau_3 + m_4^2 \rho_4 \tau_4 - 2m_3 m_4 \vec{j}_3 \cdot \vec{j}_4) + \mathcal{V}(\rho_3, \rho_4),$$

(A1)

where the kinetic energy $\tau_q$ and current $\vec{j}_q$ densities are defined in Ref. 13, and the functions $g_3$, $g_4$ and $g_{34}$ are not identically zero if the atoms have an effective mass $m_q^*$ different from the bare mass (we have taken $\hbar = 1$ thorough the Appendix). In the static case, the system is invariant under time-reversal and the currents vanish. This allows to construct the $g$ functions from the static density functional without having to reparametrize it to reproduce the basic thermodynamical properties of the mixture. Yet, casting the static density functional into a form like Eq. (A1) leaves plenty of freedom, as it is easy to convince oneself. The grand potential density is then built from the energy density, the chemical potentials and particle densities: $\omega = \mathcal{E} - \sum_q \mu_q \rho_q$.

One then applies to the $^3\text{He}$ and $^4\text{He}$ fluids a radial collective velocity $\vec{u}_q(\vec{r}, t)$ ('boost') deriving from velocity potential fields $s_q(\vec{r}, t)$, $\vec{u}_q(\vec{r}, t) = \nabla s_q(\vec{r}, t)$ and takes into account how the particle, kinetic energy and current densities are affected by these boosts. If $\vec{j}_3 = \vec{j}_4 = 0$ in the static system, it is easy to check that the imaginary-time Lagrangian can be written as

$$\mathcal{L} = \sum_q \left[ m_q \dot{\rho}_q s_q - \frac{1}{2} m_q \rho_q (\nabla s_q)^2 \right] - \frac{1}{2} G_{34} (\nabla s_3 - \nabla s_4)^2 + [\omega(\rho_q, \tau_q) - \omega(\rho_{qm}, \tau_{qm})],$$

(A2)

where $\frac{1}{2} G_{34} \equiv m_3^2 m_4^2 \rho_3 \rho_4 g_{34}$. The imaginary-time Hamiltonian $\mathcal{H}$ is readily obtained from $\mathcal{L}$:

$$\mathcal{H} = \sum_q m_q \rho_q (\nabla s_q)^2 + \frac{1}{2} G_{34} (\nabla s_3 - \nabla s_4)^2 - [\omega(\rho_q, \tau_q) - \omega(\rho_{qm}, \tau_{qm})],$$

(A3)

and then the Hamilton equations

$$m_q \dot{\rho}_q = \frac{\delta \mathcal{H}}{\delta s_q}$$

$$m_q \dot{s}_q = - \frac{\delta \mathcal{H}}{\delta \rho_q},$$

(A4)

yield, respectively, the continuity and equations of motion:

$$m_3 \dot{\rho}_3 = - m_3 \nabla (\rho_3 \vec{u}_3) - \nabla [G_{34}(\vec{u}_3 - \vec{u}_4)]$$

$$m_4 \dot{\rho}_4 = - m_4 \nabla (\rho_4 \vec{u}_4) + \nabla [G_{34}(\vec{u}_3 - \vec{u}_4)]$$

(A5)
\[
m_3 \frac{d\vec{u}_3}{dt} = -\nabla \left\{ \frac{1}{2} m_3 \vec{u}_3^2 + \frac{\partial G_{34}}{\partial \rho_3} (\vec{u}_3 - \vec{u}_4)^2 - \frac{\delta \omega}{\delta \rho_3} \right\},
\]
\[
m_4 \frac{d\vec{u}_4}{dt} = -\nabla \left\{ \frac{1}{2} m_4 \vec{u}_4^2 + \frac{\partial G_{34}}{\partial \rho_4} (\vec{u}_3 - \vec{u}_4)^2 - \frac{\delta \omega}{\delta \rho_4} \right\}.
\]

(A6)

The above equations are quite similar to those obtained in Ref. 46 to describe nuclear collective vibrations, and at this point it is appropriate to recall the last paragraph of Sect. IV. Linearizing Eqs. (A5,A6) is straightforward and proceeds as outlined in Ref. 32. We do not write down the resulting equations, which are more involved than those given in the above reference because here we have kept in the equations the terms arising from the possible existence of a \(g_{34}\) contribution. Rather, we finish the Appendix with a short discussion about possible choices for the \(g\) functions.

The more obvious and simple one is to take \(g_4 = g_{34} \equiv 0\) and \(g_3 \rho_3 \equiv 1/(2m_3^*)\), where \(m_3^*(\rho_3, \rho_4)\) is the \(^3\)He effective mass in the mixture. This leads to linearized equations to determine \(T^*\) which are extremely cumbersome but still manageable. They have also been employed to obtain the density functional results presented in this work, and consistent with this choice is to write the collective kinetic energy in the capillarity approximation as indicated by Eq. (22).

The more general expression \(g_3 \rho_3 + m_4^2 \rho_4 g_{34} \equiv 1/(2m_3^*)\) holds if one uses the accurate parametrization.

\[
\frac{1}{2m_3^*} = \frac{1}{2m_3} \left[ 1 - \frac{\rho_3}{\rho_{3c}} - \frac{\rho_4}{\rho_{4c}} \right]^2,
\]

(A7)

where the fitting constants \(\rho_{3c}, \rho_{4c}\) can be found in the above references. It can be checked that setting \(g_4 = 0\) and splitting the above relation into

\[
g_3(\rho_3, \rho_4) = \frac{1}{2m_3} \left[ \frac{\rho_3}{\rho_{3c}} - \frac{2}{\rho_{3c}} + \frac{2\rho_4}{\rho_{3c} \rho_{4c}} \right],
\]

\[
g_{34}(\rho_3, \rho_4) = \frac{1}{2m_3 m_4^2} \left[ \frac{\rho_4}{\rho_{4c}^2} - \frac{2}{\rho_{4c}} \right],
\]

(A8)

the good fit to the experimental \(m_3^*\) is retained, and

\[
\frac{m_4}{m_4^*} = 1 + \frac{m_3}{m_4} \left[ \frac{\rho_4}{\rho_{4c}^2} + \frac{2\rho_3}{\rho_{3c} \rho_{4c}} - \frac{2}{\rho_{4c}} \right] \rho_3,
\]

(A9)

which yields \(m_4^*/m_4 = 1.31\) for one \(^4\)He impurity in normal liquid \(^3\)He at saturation. This value is in agreement with the experimental one.
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FIGURES

FIG. 1. Schematic barrier $\Delta \Omega(\delta)$ and inverted barrier well.

FIG. 2. Schematic figure to illustrate the calculation of the collective mass.

FIG. 3. $T^*$ (mK) as a function of $\Delta x(\%)$. Dashed lines are the results obtained within the capillarity approximation, and solid lines are the results of a density functional plus FIA calculation. The upper solid and dashed curves correspond to $P = 1$ bar, and the lower ones to $P = 0$.

FIG. 4. Density profile of the critical nucleation configuration obtained using either a density functional method (thick lines), or the capillarity approximation (thin lines). The values correspond to $P = 0$ and $\Delta x = 16\%$.
\[ \rho(r) \]

- \( \rho_{30} \)
- \( \delta \)
- \( R_c \)
- \( \rho_{3m} \)
- \( \rho_{4m} \)

- \( r \)
The graph shows the density function $\rho(r)$ as a function of $r$. Two curves are depicted, labeled $\rho_3$ and $\rho_4$. The curve $\rho_3$ decreases as $r$ increases, while $\rho_4$ increases. The value of $\rho_4$ at $r = R_c$ is indicated, where $R_c$ is a specific radius marked on the graph.