Thermal nucleation of cavities in liquid helium at negative pressures

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We have investigated the nucleation rate at which cavities are formed in ^{4}He and ^{3}He at negative pressures due to thermal fluctuations. To this end, we have used a density functional that reproduces the He liquid-gas interface along the coexistence line. The inclusion of thermal effects in the calculation of the barrier against nucleation results in a sizable decrease of the absolute value of the tensile strength above 1.5 K.

Theoretical investigations of liquid-helium properties at negative pressures[1–4] have been prompted by recent experiments carried out by Nissen *et al.*[5] and by Xiong and Maris[6] using ultrasonic waves. This method allows the study of cavitation in very small liquid volumes, considerably avoiding the possibility of heterogeneous nucleation at electron bubbles.

Although the experimental results reported in Ref. 5 for ^{4}He at temperatures above 1.5 K seemed to be well reproduced by classical nucleation theory (ClNT),[8] the experiment carried out in Ref. 6 appears to discard this possibility. A serious argument against the interpretation of the experimental findings of Ref. 5, already raised in Ref. 6 and confirmed in Ref. 4, is that the critical pressure P_c at which liquid ^{4}He becomes *macroscopically* unstable is bigger than the tensile strength yielded by ClNT and by the experiment reported in Ref. 5. (To avoid any possible misunderstanding, here we define the tensile strength as a negative quantity.)

Xiong and Maris have found that the tensile strength for nucleation of bubbles in ^{4}He for temperatures in the $0.8 - 2$ K range, is ~ -3 bars. To analyze their experimental results, they have resorted to a method that represents a considerable improvement over the ClNT. It is based on a density functional (DF) whose free parameters are fixed to yield the experimental velocity of sound propagation in the liquid as a function of the density ρ, and includes a gradient term $\lambda(\nabla\rho)^2$ adjusted so as to reproduce the surface tension of ^{4}He at $T = 0$ K.

Using their revised nucleation theory, they have found a tensile strength that goes from ~ -9 bars at $T = 0$ K, to ~ -6.5 bars at $T = 2$ K, still lying in absolute value well above their experimental data. There may be several reasons for this disagreement. The first is the validity of their functional in the density domain corresponding to negative pressures. However, since other equations of state[4] (see also below) yield very similar values for P_c (around ~ -9 bars at $T = 0$ K), we do not believe this to be the cause of the disagreement. If the ultrasonic technique used in these experiments discards the possibility of heterogeneous nucleation, nucleation on vortice lines is likely the main origin of the discrepancy. Xiong and Maris[6] have estimated the critical pressure P_c^v for nucleation due to vortices to be ~ -6.5 bars at $T = 0$ K whereas Dalfovo,[9] using a nonlocal DF, has obtained a value of ~ -8 bars, much closer to P_c.

In this work, we want to address the effect that a nonzero temperature has on the nucleation barrier. This has been overlooked in all previous calculations and is of relevance in order to put on a firmer basis which part of the disagreement between theory and experiment can be attributed to nucleation of bubbles on a vortice line in the case of ^{4}He. For ^{3}He, our results constitute the first detailed study of the tensile strength using a realistic DF, and can be of some relevance in view of the planned experiments on this helium isotope.[6]

Our starting point is the following free energy functional:[10,11]

$$f(\rho,T) = f_{\rm ni}(\rho,T) + \tfrac{1}{2}b\rho^2 + \tfrac{1}{2}c\rho^{2+\gamma} + \beta\frac{(\nabla\rho)^2}{\rho} + \xi(\nabla\rho)^2 \ , \quad (1)$$

where $f_{\rm ni}$ is the free energy density of a noninteracting Fermi or Bose gas, and the parameters b, c, γ, β, and ξ have been adjusted so as to reproduce physical properties of the homogeneous liquid and of the liquid-gas interface. We want to point out that the surface tension as

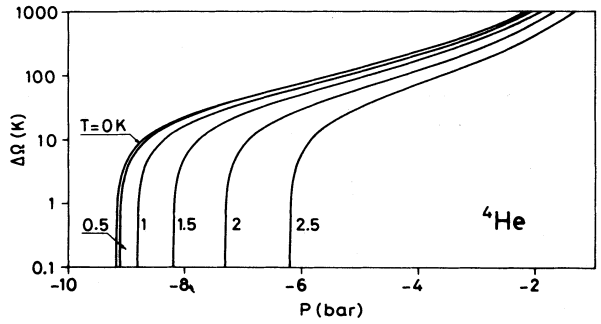

FIG. 1. Nucleation barrier $\Delta\Omega$ for ^{4}He as a function of pressure for several temperatures T.

a function of T, the isotherms, and the vapor density in equilibrium with the liquid phase are well reproduced by our model up to temperatures above the ones of interest for the present study. We refer the reader to Refs. 10 and 11 for a detailed discussion of the DF (1) and the results obtained from its application to the study of liquid-gas equilibrium.

At a given T, the density profile of a bubble is obtained solving the Euler-Lagrange equation

$$\frac{\delta f}{\delta \rho} = \frac{\partial f}{\partial \rho} - \nabla \frac{\partial f}{\partial \nabla \rho} = \mu \ , \tag{2}$$

where μ is the chemical potential corresponding to any density ρ_m such that the pressure of the homogeneous liquid $P(\rho_m, T)$ is negative, corresponding to a metastable state. The boundary conditions for finding a physical solution to (2) are $\rho'(0) = 0$ and $\rho(r \to \infty) = \rho_m$. Finally, the nucleation barrier is obtained from the difference between the grand potential of the bubble and of the homogeneous system:

$$\Delta\Omega = \int d\mathbf{r}\,[f(\rho, T) - f(\rho_m, T) - \mu(\rho - \rho_m)] \, . \tag{3}$$

This procedure yields $\Delta\Omega$ as a function of P and T. Figures 1 and 2 show the barrier $\Delta\Omega$ for several temperatures in the case of ^{4}He and ^{3}He, respectively. The inclusion of thermal effects in the calculation lowers $\Delta\Omega$ because the surface tension decrease with increasing T. Moreover, the density inside the bubble increases, thus

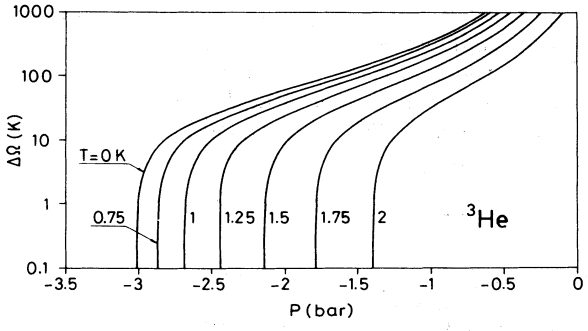

FIG. 2. Nucleation barrier $\Delta\Omega$ for ^{3}He as a function of pressure for several temperatures T.

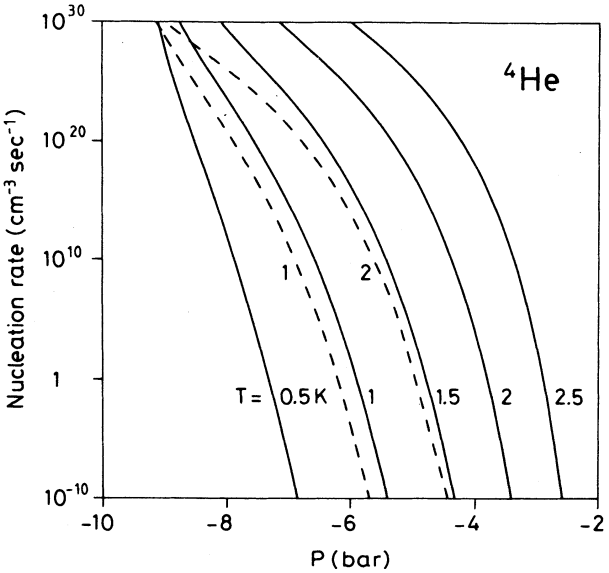

FIG. 3. Thermal nucleation rate of bubbles in ^{4}He as a function of pressure for several temperatures T. The dashed lines represent the nucleation rates obtained from the $\Delta\Omega(T = 0)$ barrier.

making its structure more similar to the homogeneous metastable phase.

To calculate the nucleation rate Γ_T due to thermal fluctuations as a function of P and T we have used[12]

$$\Gamma_T = \Gamma_T^0 \exp\left(-\frac{\Delta\Omega}{kT}\right) \ . \tag{4}$$

The prefactor Γ_T^0 depends on the liquid characteristics and on the dynamics of the cavity formation process.[7] For the sake of simplicity, we will use the same prefactor as Xiong and Maris,[2] i.e., $\Gamma_T^0 = kT/(hV_N)$, where V_N is

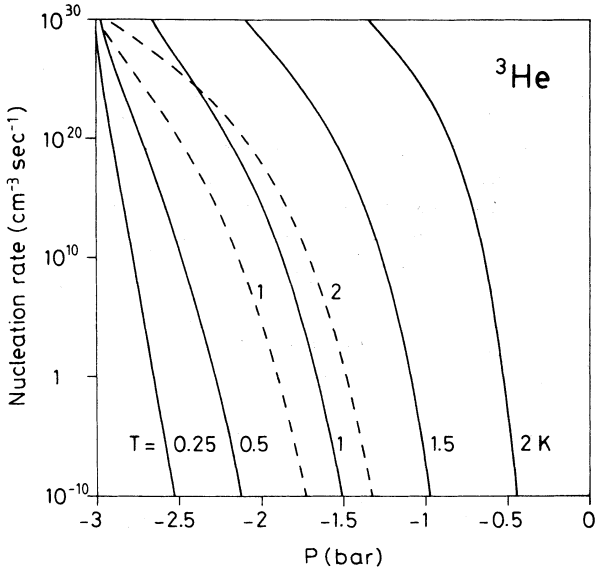

FIG. 4. Same as Fig. 3 for ^{3}He.

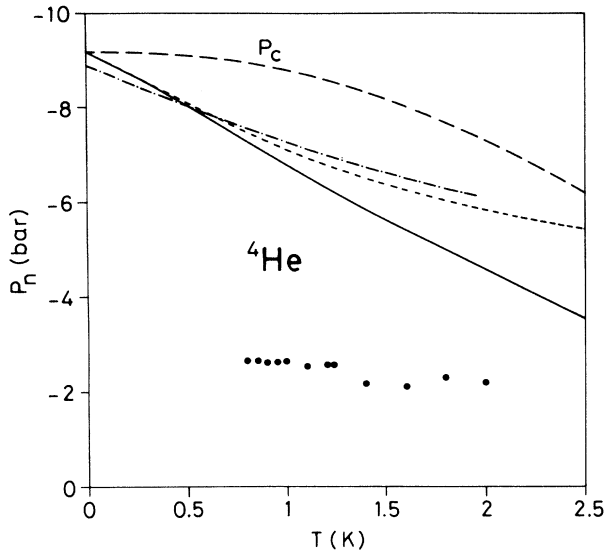

FIG. 5. Tensile strength P_n needed to cause nucleation in ^{4}He corresponding to a product of experimental volume and time equal to 2.5×10^{-13} cm^3 sec. The dash-dotted line and dots are the theoretical and experimental results of Ref. 6, respectively. The thick line has been obtained from the $\Delta\Omega(T)$ barrier, and the short-dashed line from the $\Delta\Omega(T=0)$ barrier. The spinodal line $P_c(T)$ is represented by a long-dashed line.

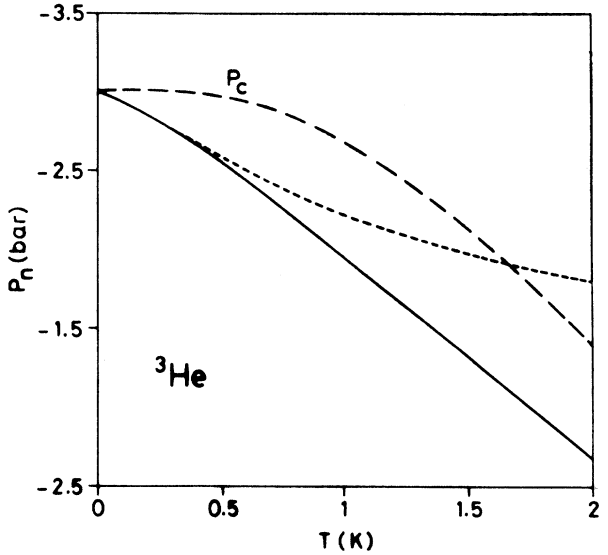

FIG. 6. Same as Fig. 5 for ^{3}He.

the volume of a sphere of radius 10 Å representing the critical bubble.

Figures 3 and 4 show Γ_T for ^{4}He and ^{3}He. The dashed lines correspond to nucleation rates calculated from the $\Delta\Omega(T=0)$ barrier. One can see that a better estimate of $\Delta\Omega(T)$ increases Γ_T by more than one order of magnitude at pressures relevant for bubble nucleation.

We have obtained the tensile strength P_n solving the equation

$$V\tau\,\Gamma_T(P_n) = 1 \ , \tag{5}$$

where the experimental volume V and time τ have been taken from Ref. 6 ($V\tau = 2.5 \times 10^{-13}$ cm^3 sec) to allow for a sensible comparison with their results.

The tensile strength P_n for ^{4}He and ^{3}He is shown in Figs. 5 and 6, respectively. The short-dashed line has been obtained from $\Delta\Omega(T=0)$, and the small difference between this curve and Xiong-Maris results (dash-dotted line) for ^{4}He is due to the different DF used in both calculations. For comparison, we also show the critical pressure $P_c(T)$ (long-dashed line) at which liquid He becomes macroscopically unstable (spinodal line). It is worth it to remark that the use of $\Delta\Omega(T=0)$ to obtain Γ_T constitutes a poor approximation at moderated temperatures, yielding a tensile strength bigger in absolute value than $\mid P_c \mid$ at $T \sim 1.5$ K for ^{3}He and at $T \sim 2.5$ K for ^{4}He.

Our calculations show that $\mid P_n \mid$ decreases appreciably at temperatures above 1.5 K (we recall that the liquid-gas critical temperatures are 3.32 K for ^{3}He and 5.20 K for ^{4}He). Yet, the discrepancy with the experimental data[6] (dots in Fig. 5) is large, of the order of 2 bars at $T = 2$ K.

To test the improved nucleation theory, ultrasonic experiments on ^{4}He are called for above the λ temperature $T_\lambda = 2.17$ K to get rid of the influence of bubble nucleation on vortices. Above T_λ, one should then expect the experimental $\mid P_n \mid$ to increase, thus improving the agreement between theory and experiment. Alternatively, experiments on cavitation in ^{3}He could also help understand if the existing discrepancy for ^{4}He is still due to shortcomings of the nucleation theory, to a sizable nucleation on vortice lines, or to an undetected effect.

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[12]It has been shown in Ref. 2 that for ^{4}He, quantum tunneling through the nucleation barrier can be safely neglected for temperatures above ~ 0.3 K.