Quantum Cavitation: a comparison between superfluid helium-4 and normal liquid helium-3

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Cavitation has now been studied in superfluid helium-4 and in normal liquid helium-3, both theoretically and experimentally. We compare the two cases and discuss the existence of a crossover from quantum cavitation, where bubbles are nucleated by tunneling, to classical cavitation where their nucleation is thermally activated. In helium-3, where evidence for quantum nucleation is lacking, the interpretation of the experimental results leads to two related questions. The first one concerns the extrapolation of the properties of this Fermi liquid at negative pressure. The second one concerns the validity of present theories of quantum cavitation in a Fermi liquid.

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1. INTRODUCTION

Cavitation is the nucleation of bubbles in a liquid. It is a problem which one tries to avoid in hydraulics, and is also the physical mechanism which is used in an ultrasonic cleaning bath to remove dust particles from a surface. Useful or not, cavitation is a phenomenon which is hard to control in ordinary situations because the nucleation of bubbles is "heterogeneous", meaning that it takes place on impurities, solid particles, gaseous inclusions or walls, so that it is not an intrinsic property of the liquid.

There are several reasons to study cavitation in liquid helium. In principle, cavitation is simpler to understand in liquid helium because this liquid can be prepared free from impurities. Furthermore, liquid helium is a simple liquid whose equation of state over a wide pressure range has been calculated by several complementary methods.[1,7–9] Finally, one can compare two different helium liquids in the same temperature range (40 to 1000 mK), superfluid helium-4 and normal liquid helium-3. Liquid helium-3 is lighter, has less internal cohesion and has very different quantum properties (it is a viscous Fermi liquid except at ultralow temperatures where it also becomes superfluid).

By quantum cavitation we refer to the process in which the nucleation of bubbles takes place by quantum tunneling. This is in contrast to classical cavitation, where the nucleation is thermally activated over an energy barrier. Maris[1] and Guilleumas *et al.*[2] predicted that quantum cavitation should occur both in superfluid helium-4 and in normal liquid helium-3 at low enough temperature (below 200 mK in helium-4 and 120 mK in helium-3) and at large negative pressures (around - 9 bars and - 3 bars, respectively). Studies of quantum cavitation in liquid helium probe the extreme limit of metastability of a simple liquid at negative pressure and near $T = 0$. It is also an interesting approach to the investigation of macroscopic quantum phenomena.

Our experimental set-up is already described elsewhere.[3–5] The experimental cell is attached to the mixing chamber of a dilution refrigerator with optical access. Inside this cell, a hemispherical piezoelectric ceramic is used to produce bursts of 1 MHz ultrasound which are focussed into a region of about half the acoustic wavelength ($\lambda/2 \approx 100$ μm) far from any wall (the transducer wall is 8 mm away). A laser beam is also focussed through the windows of our cryostat onto the acoustic focus. Light is scattered both by the acoustic wave and by the bubbles. At the acoustic focus, the acoustic intensity is very large, several bars in pressure and typically 30% in density. In our last experiments, we used sound bursts of duration only 6 μs, i.e., only six oscillations at 1 MHz, together with a repetition rate as low as 0.2 Hz. This resulted in a dissipation in the cell as low as 2 μW, and consequently we were able to work at temperatures down to 40 mK.

Figure 1 shows two typical signals recorded in helium-3 by Caupin *et al.*[5] The two traces have been obtained with exactly the same sound intensity; the lower one is a recording of the light scattered by the acoustic wave only, when no cavitation occurs. On the upper one, a large additional scattering is recorded which corresponds to the scattering by the bubble. As already observed in helium-4, the collapse of this bubble at time t = 150 μs leads to the nucleation of a smaller bubble. Below a certain threshold in sound amplitude, all signals are exactly like the lower trace, i.e., there is no cavitation. Above the threshold, the trace varies from sound pulse to pulse because bubbles have a distribution in birth times, and consequently different energy, maximum size, and total life time. In the vicinity of the

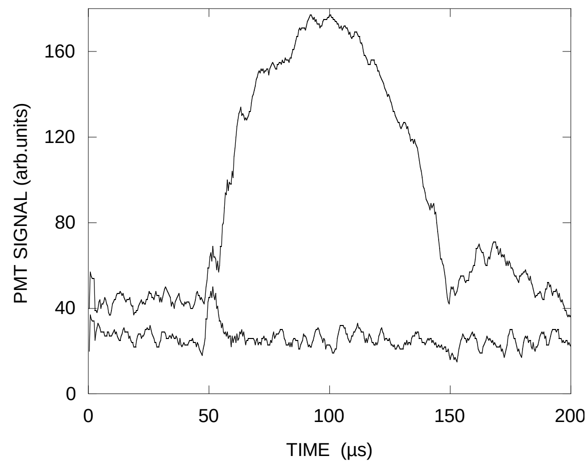

Fig. 1. Two recordings of the scattered light detected by the photomultiplier tube. The excitation is the same; only the upper recording shows cavitation; the lower one shows a small light scattering from the acoustic wave only.

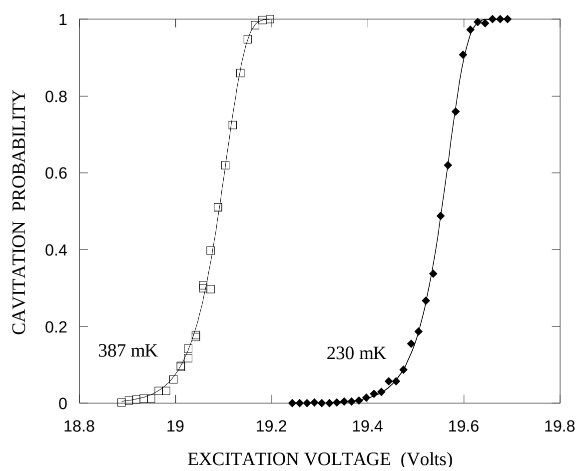

Fig. 2. Cavitation probability versus excitation voltage at $P_{stat} = 20\,\mathrm{mbars}$ and two different temperatures: $T = 230\,\mathrm{mK}$ and $T = 387\,\mathrm{mK}$. The solid lines are fits with Eq. (3).

threshold, cavitation is clearly stochastic, with some traces showing bubble nucleation and others not.

We measured the nucleation probability Σ by counting for a series of several hundred sound pulses the number of times at least one bubble was produced. As shown in Fig. 2, the cavitation probability increases smoothly from 0 to 1 in a narrow voltage interval. The plot of Σ versus voltage always has an asymmetric S-shape, and this has a simple explanation as outlined below.

2. THEORETICAL BACKGROUND

Let us start with the standard classical theory of nucleation. In the so called "thin wall approximation", one assumes that the nucleus of the gas phase is a bubble of radius R with the same surface tension α as the bulk liquid. The free energy of such a bubble is[6]

$$F(R) = 4\pi R^2 \alpha + \frac{4}{3}\pi R^3 P \tag{1}$$

where P is the negative pressure in the liquid (we neglect the saturated vapor pressure which is exponentially small for liquid helium at low temperature). $F(R)$ has a maximum for a certain critical radius $R_c = 2\alpha/|P|$. The energy of this critical nucleus is the energy barrier for nucleation $E = 16\pi\alpha^3/3P^2$. The nucleation rate per unit time and per unit volume is then $\Gamma_0 \exp(-E/k_B T)$, where Γ_0 is an attempt frequency per unit volume. Thus, the cavitation probability is

$$\Sigma = 1 - \exp\left[-\Gamma_0 V_{\mathrm{exp}} \tau_{\mathrm{exp}} \exp\left(-E/k_B T\right)\right], \tag{2}$$

where $V_{\mathrm{exp}}\tau_{\mathrm{exp}}$ is the product of the experimental time and the experimental volume. An equivalent way to write the above equation is:

$$\Sigma = 1 - \exp\left\{-\ln 2 \exp\left[\left(\frac{V}{V_c} - 1\right)\xi\right]\right\}, \tag{3}$$

which is easy to obtain by expanding E around the cavitation threshold V_c, where $\Sigma = 0.5$ and the barrier is E_c. The coefficient $\xi = (E_c/k_B T)|d\ln E/d\ln V|$ is a dimensionless quantity measuring the inverse width of the curve. The asymmetric S-shape in Fig. 2 is characteristic of the double exponential in the above equations. Thanks to various improvements in the stability of our experiment, the quality of the fit with Eq. (3) is even better in helium-3 than previously obtained in helium-4; it provides strong evidence that we do not miss bubbles, and that we are really looking at a metastable state whose decay rate varies exponentially with the control parameter, i.e., the voltage V that governs the amplitude of the emitted acoustic wave.[4]

Of course, the standard nucleation theory is oversimplified and needs to be improved. First, the magnitude of the critical radius is about 10 Å, i.e., comparable to the thickness of the macroscopic liquid-gas interface, so that the thin wall approximation is not valid and a density functional theory has to be used to optimize the density profile of the nucleus and calculate the activation energy. Maris[7] used the simple density functional:

$$F = \int [f(\rho) + \lambda(\bigtriangledown\rho)^2]dV, \tag{4}$$

and adjusted λ to obtain the right value of the surface tension α.

Secondly, a good theory has to account for the existence of the "spinodal limit". The above standard theory predicts that, as T tends to zero, the activation energy corresponding to a probability 0.5 should tend to zero; as a consequence, the cavitation threshold pressure should diverge towards $-\infty$, and this is not possible. Indeed, since the intermolecular attraction decreases with distance, there is a maximum stress which a liquid can stand. It corresponds to a lower bound in density below which the liquid is totally unstable with respect to the formation of the gas phase. This extreme limit of possible metastability is the spinodal limit $(P_{\rm sp}, \rho_{\rm sp})$. At this point on the equation of state $P(\rho)$, the compressibility is infinite and the sound velocity vanishes. Maris[1,7] has proposed the equation of state

$$P - P_{\rm sp} = \frac{b^2}{27}\,(\rho - \rho_{\rm sp})^3, \tag{5}$$

where b is a constant. This gives a sound velocity

$$c = \frac{b}{3}(\rho - \rho_{sp}). \tag{6}$$

By a suitable choice of b, $P_{\rm sp}$, and $\rho_{\rm sp}$ a remarkably good fit to sound velocity measurements for positive pressure can be achieved.[7] The results for the spinodal are

$$P_{\rm sp} = -9.52 \text{ bar}, \quad \rho_{\rm sp} = 0.095 \text{ g cm}^{-3}, \tag{7}$$

in helium-4 (to be compared to ρ_0=0.1451 g cm^{-3} at $P = 0$), and

$$P_{\rm sp} = -3.09 \text{ bar}, \quad \rho_{\rm sp} = 0.054 \text{ gcm}^{-3}, \tag{8}$$

in helium-3 where ρ_0=0.0815 g cm^{-3} at $P = 0$. In helium-4, Dalfovo[8] used a more elaborate form of density functional and found the same spinodal pressure with a somewhat higher spinodal density (0.105 g cm^{-3}). Boronat[9] used a Monte Carlo method and also found similar values (-9.3 bar and 0.105 g cm^{-3}). We thus believe that the spinodal limit in liquid helium is rather well established theoretically. In the density functional method, the existence of the spinodal limit is, of course, included in the function $f(\rho)$. One should note that the existence of a spinodal limit has an important effect on the calculation of the energy barrier E, but it does not change the double exponential form of the cavitation probability.

Finally, another important improvement has to be made to the standard theory: one has to allow for the possibility of quantum tunneling through the energy barrier. The probability for quantum cavitation obeys an equation very similar to the classical one:[1,2] the exponent E/k_BT is simply replaced by the action $B/\hbar$. To calculate the action B is a difficult and interesting problem. Maris[1] did it by using the inverted potential method.[10] Dissipation is neglected, and the "bounce" trajectory is optimized numerically to minimize B. This calculation is done at $T = 0$ and uses nothing but the equation of state $P(\rho)$ and the parameter λ. An estimate of the attempt frequency finally leads to the probability for quantum cavitation, which is compared to the classical, thermally activated, one. A crossover temperature T^* is deduced, at which the two probabilities are equal. Maris found $T^* =$ 200 mK in helium-4 and 120 mK in helium-3. The calculation by Guilleumas *et al.* includes tunneling at finite temperature and leads to similar values of T^*.

3. QUANTUM CAVITATION: HELIUM-4 VERSUS HELIUM-3

In order to check the prediction of such a crossover, we first studied cavitation as a function of temperature in superfluid helium-4. As shown in Fig. 3, the cavitation probability is independent of T for low enough temperature in helium-4. We defined the cavitation threshold as the voltage V_c where the probability is 0.5, and studied $V_c(T)$. Fig. 4 shows this temperature variation. V_c goes through a maximum around 0.75 K, which at first sight is surprising, but is easy to explain. The ceramic emits a sound wave which propagates over a distance of 8 mm before reaching the focus. It happens that at 0.75 K the phonon-roton collision time becomes equal to the inverse sound frequency, and consequently there is a large peak in the ultrasonic attenuation.[11] As a consequence, in order to obtain a constant wave amplitude at the focus, one has to increase the excitation voltage to compensate for this attenuation. In the case of helium-4, we were able to verify this attenuation by measuring the scattering of light by the sound wave in the focal region. We determined the correction to be applied to the measured V_c and obtained the "corrected data" as shown in the figure. After

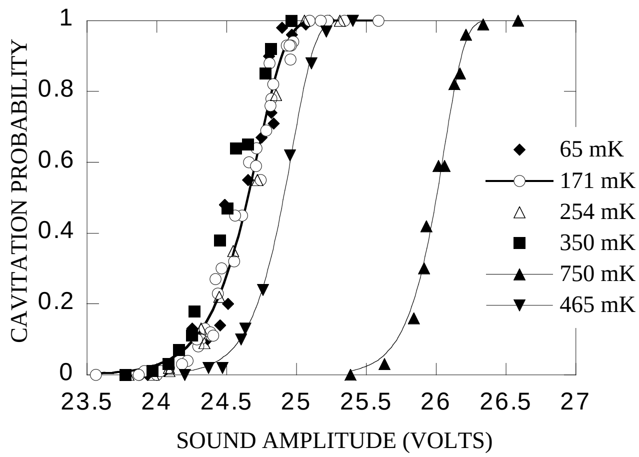

Fig. 3. Cavitation probability in helium-4 as a function of the voltage applied to the transducer for a series of different temperatures. The cavitation threshold is defined as the voltage corresponding to a probability 0.5.

this correction, the cavitation threshold $V_c(T)$ shows a well defined plateau below 0.6 K, followed by the decrease with increasing temperature which is expected for a classical, thermally activated, nucleation process.

The observation of a temperature-independent cavitation which is stochastic in a finite range of the control parameter is strong evidence for quantum cavitation. Indeed, if quantum cavitation did *not* occur, in the $T = 0$ limit cavitation would become deterministic. Thus, if the system did not reach the spinodal, the energy barrier would be non-zero and in the absence of any thermal fluctuations cavitation could never occur, so that the cavitation probability would be zero, whereas if the spinodal were reached, bubbles would appear with a 100 % probability. Thus, the probability as a function of voltage would show a sharp step. On the contrary, Lambaré *et al.*[4] observed that the probability curve keeps a constant, temperature independent width, and is temperature independent at low T. In addition, Lambaré *et al.*[4] were able to show that the width of the transition was in reasonable agreement with theory. It was estimated that the threshod was at -9.23 bars, about 0.3 bars above the spinodal. At this pressure the action is about $36\hbar$, the energy barrier is 10.5 K, and the sound velocity has decreased to 75 m s^{-1} from the zero pressure value of 238 m s^{-1}. The disagreement between the theoretical crossover temperature (200 mK), and the measured T^* (600 mK), is only apparent. Since the acoustic wave is adiabatic (at least in a first approximation), the pressure wave is accompanied by a temperature oscillation, which maintains a constant entropy. In superfluid helium-4,

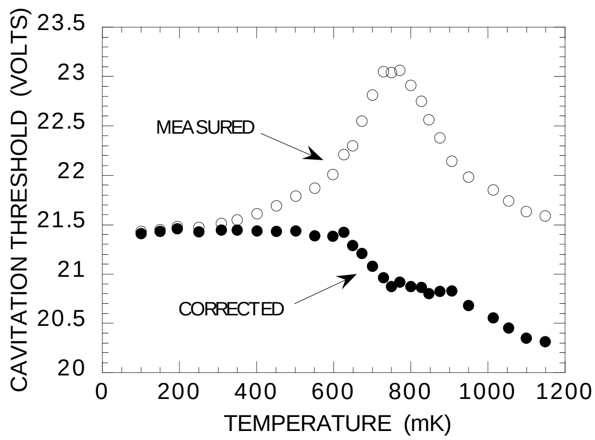

Fig. 4. Cavitation threshold versus temperature in helium-4. The raw data show a maximum at 750 mK which is due to the temperature variation of the sound attenuation. After correction for this effect, the temperature independent plateau apparently extends up to $T < 0.6\ K$). This is consistent with a crossover temperature T* = 0.2K from quantum cavitation to classical cavitation (see text)

the entropy is dominated by phonons at low T, so that it is proportional to $(T/c)^3$. As a consequence, if the pressure swing at the focusis sufficiently negative to reduce the sound velocity by a factor 3, the local instantaneous temperature will be reduced by the same factor. Thus, although the *static* temperature at the crossover is found to be 0.6 K, the instantaneous temperature at the peak negative pressure is 0.2 K, consistent with the theoretically predicted T^*.

Of course, our claim that we have observed quantum cavitation in helium-4 would be strongly supported by an accurate measurement of the time evolution of P, ρ, and T at the acoustic focus. Before attempting this type of measurement, however, we have compared cavitation in helium-4 with helium-3 in the same low temperature limit. Fig. 5 shows the temperature variation of the cavitation threshold in helium-3. Figures 5 and 6 show data for the cavitation threshold, after application of a correction to account for the small but finite attenuation of sound waves, and for some small temperature gradients in the cell. The corrected data can be fit very well by a $T^{3/2}$ law over the entire temperature range, i.e., there is no evidence for a transition from the $T^{3/2}$ behavior to a temperature-independent regime. From this observation, Caupin *et al.* concluded that they had “not yet found any evidence for a crossover toward cavitation by quantum tunneling”.[5]

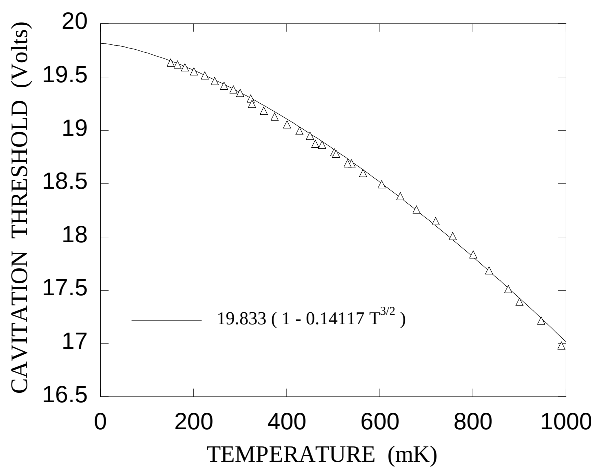

Fig. 5. Cavitation threshold versus temperature in helium-3 at $P =$ 20 mbars up to 1000 mK. The solid line is a best fit with a power law through the data.

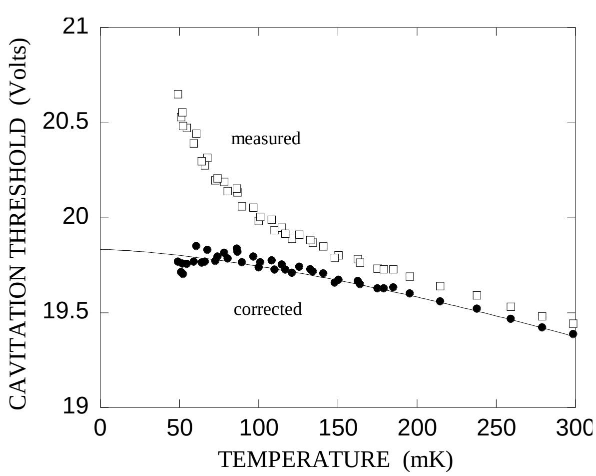

Fig. 6. Cavitation threshold versus temperature in helium-3 at $P =$ 20 mbars below 300 mK. The solid line is the same as on Fig. 5. Black circles were obtained by Caupin et al. after correcting their raw data (open squares) from the effects of a small sound attenuation and a small temperature gradient in their cell.

One possibility is that the instantaneous temperature is different from the measured static temperature, as in the case of helium-4. In helium-3 also, the effect of dissipation is small, so that the sound wave is adiabatic in first approximation. For a degenerate Fermi liquid, the entropy per mole is equal to the specific heat

$$S = C = \frac{\pi^2}{2}\frac{m^*}{m}\frac{Rk_BT}{\epsilon_F} \propto \frac{m^*}{m}\frac{1}{\rho^{2/3}}. \tag{9}$$

Stringari[13] noticed that in helium-3 the square root of the inverse effective mass is linear in the density. We have confirmed that Greywall's data[14] are very well described by

$$(m/m^*)^{1/2} = 1.0166(1 - 5.138\rho), \tag{10}$$

with ρ in cgs units. Thus, we expect the entropy to vary with density and temperature according to

$$S \propto \frac{1}{(1-5.138\rho)^2}\frac{1}{\rho^{2/3}}T. \tag{11}$$

Such a law predicts that there should be an adiabatic warming of 14% for a pressure swing from $P = 0$ to -3.1 bar. As a consequence, the crossover to quantum cavitation should be observed for a cell temperature of about 100 mK, instead of 120 mK. It is not possible to conclude from the data of Fig. 6 whether or not there is a transition to a temperature-independent V_c at around 100 mK.

4. SOME CONSIDERATIONS REGARDING THE THEORY OF QUANTUM CAVITATION IN HELIUM-3

Caupin *et al.* point out that more accurate data at lower temperatures may show a crossover to a quantum regime, but it is interesting to consider other hypotheses. One possibility is that superfluidity is necessary for the quantum nucleation of bubbles. Of course, the formation of the critical nucleus involves about 1000 atoms, and when considering the representation of such a number of particles by a single wavefunction extending through the energy barrier in configuration space, one could consider that some kind of phase coherence is needed which has to be provided by the superfludity. However, we believe that the necessary coherence is nothing more than the coherence in a density wave.

Still, there is another reason why helium-3 may be different from helium-4, namely the special properties of a Fermi liquid. The isothermal and adiabatic sound velocities, c_T and c_S are defined by

$$c_T^2 = \left(\frac{\partial P}{\partial \rho}\right)_T , \qquad \text{and} \qquad c_S^2 = \left(\frac{\partial P}{\partial \rho}\right)_S . \tag{12}$$

The isothermal sound velocity is not directly measurable in an experiment. The adiabatic sound velocity governs the propagation of sound waves of frequency ω such that $\omega\tau < 1$, where τ is the collision time between the helium-3 quasiparticles. For $\omega\tau > 1$ the sound velocity is c_0, the zero sound velocity. The relaxation time τ varies with temperature as

$$\tau = 2 \times 10^{-12}/T^2 \quad \text{s.} \tag{13}$$

The zero sound velocity c_0 is larger than c_S by an amount which varies with pressure, and is approximately 4 % at $P = 0$. c_S is larger than c_T, but the difference tends to zero in the limit $T \to 0$.

It is the isothermal sound velocity which goes to zero at the spinodal. The energy barrier E for classical nucleation is defined as the least amount of free energy that is needed in order to pass from the uniform liquid to a state of lower energy containing a large bubble. Since this process can be carried out isothermally, this energy clearly goes to zero as $P \to P_{\mathrm{sp}}$. Now consider the quantum nucleation process near to this pressure. In quantum nucleation it is necessary to minimize the action B for the time that the system takes to pass under the barrier. The action is defined as

$$B = \frac{2}{\hbar} \int_{t_1}^{t_2} dt[K(t) + V(t)] \equiv B_K + B_V , \tag{14}$$

where the passage under the barrier begins at t_1 and ends at t_2, $K(t)$ and $V(t)$ are, respectively, the kinetic and potential energy of the liquid at time t, and B_K and B_V are the kinetic and potential contributions to the action. The action must be minimized with respect to the time τ_{passage} that the system takes for the passage; clearly a short time reduces B_V, but increases B_K because the liquid in the vicinity of the growing bubble has to move more rapidly. At the spinodal the barrier vanishes and so the time τ_{passage} can be made arbitrarily long and B_V will remain zero. In this way *both* B_K and B_V can vanish, and so the tunneling barrier too will disappear.

In the calculations of quantum tunneling performed so far,[1,2] a simple form for the free energy of the liquid has been assumed (such as Eq. 4). Since the calculations have been performed based on a zero-temperature form for the free energy, a distinction between isothermal and adiabatic sound has not been made, and is not necessary. However, it may be extremely important to consider the difference between isothermal and zero sound. As far as we are aware, there is no reason to expect that the velocity of zero sound will vanish at the spinodal. As remarked above, measurements at positive pressures show a difference between c_0 and c_T which is increasing rapidly as the pressure decreases. If c_0 remains finite at P_{sp}, then, although the static compressibility of the liquid is infinite, the liquid will have a finite stiffness for any motion that occurs with a frequency scale *omega* such that $\omega\tau > 1$. The time scale for tunneling is short compared to the quasiparticle lifetime τ, and so the effective stiffness of the liquid will be governed by c_0, rather than c_T as has been implicity assumed in the calculations performed so far. As a result there will be an increase in the potential contribution B_V to the action. This gives an increase in the minimum action B, a decrease in the tunneling probability, and as a result the crossover temperature T^* may be substantially reduced.

As the pressure approaches the spinodal, the sound velocity decreases to a value which is less than the Fermi velocity. Once this happens, there is a large increase in the ultrasonic attenuation because a sound wave can excite particle-hole pairs. As a consequence, there will be dissipation associated with the flow in the liquid around the nucleating bubble. This, too, is likely to have an effect on the tunneling rate.

While these considerations appear plausible, it is important to recognize that they are based on not much more than an extrapolation of the known properties of helium-3 into the range of negative pressures. There is a significant chance that this extrapolation is incorrect, and that the behavior of the liquid near to the spinodal shows some totally unexpected features. It is possible, for example, that at the very low density near to the spinodal in helium-3 Fermi liquid theory is inapplicable, or a very poor approximation. Clearly, more theoretical work is desirable. In addition, we are planning in the near future to make experimental measurements of the equation of state of helium in the negative pressure range.

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