Studies of Quantum Liquids in Metastable States

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Abstract. Helium can be prepared with a purity much higher than any other element. As a consequence, it is an ideal material in which to study nucleation processes. We describe work in which liquid helium has been studied at pressures below the pressure range in which the liquid is the thermodynamically stable phase. This has made possible the study of a wide range of interesting phenomena including the quantum nucleation of bubbles and the imaging of the motion of single electrons moving through the liquid.

I. Introduction

It is a great honour to be one of the recipients of Fritz London Memorial Award for LT26. I would like to thank all of the people who have helped my research over the past forty years. As a graduate student, I worked on the propagation of very high frequency sound in solids. My first involvement with helium was in 1970 when together with W.E. Massey I became interested in how sound was attenuated by interactions with phonons in liquid helium [1]. We came to the conclusion that the experimental data could only be explained if the dispersion in helium was anomalous, i.e., if the phonon dispersion relation curved upwards as the momentum increased. This was a radical idea at the time because it was in disagreement with the established theoretical ideas dating from the work of Landau and Khalatnikov in the 1950's. Fortunately, the radical idea was quickly confirmed by specific heat measurements of N.E. Phillips and his students [2] who had been reluctant to publish their results because they thought there must be something wrong in their experiment. The modification to the dispersion relation later made it possible to understand the sound attenuation, normal fluid viscosity of helium at low temperatures and some aspects of second sound propagation [3]. The details of the roton-phonon dispersion relation, in particular the fact that rotons had enough energy to evaporate helium atoms, was also important for our later work on the development of a liquid helium-based detector of solar neutrinos [4].

I became interested in metastable quantum liquids when studying the supercooling of liquid molecular hydrogen. Before describing this work, I mention some general properties of metastable states [5]. The rate of nucleation of a stable phase from a metastable phase as a result of thermal fluctuations is given by the expression

$$\Gamma_{thermal}(P,T) = \Gamma_{att,thermal} \exp(-E_{barrier}/kT), \qquad (1)$$

where $\Gamma_{att,thermal}$ is the attempt frequency per unit volume and per unit time, and $E_{barrier}$ is the height of the nucleation barrier. Nucleation can also occur as result of quantum tunnelling through the barrier. The rate of nucleation by quantum tunnelling is

$$\Gamma_{quantum}(P,T) = \Gamma_{att,quantum} \exp(-2I), \qquad (2)$$

where I is determined by the height and width of the barrier and $\Gamma_{att,quantum}$ is the attempt frequency. The total nucleation rate Γ is the sum of these two rates. This quantity can be measured by taking a volume V of the metastable phase and measuring the probability S that nucleation of the stable phase occurs. This probability is one minus the probability that nucleation does not occur within a time τ and so

$$S = 1 - \exp(-\Gamma V \tau) \,. \tag{3}$$

To measure Γ in this way it is essential to eliminate the possible effects of heterogeneous nucleation, i.e., nucleation associated with impurities or occurring on the walls of the sample container. For nucleation studies, quantum liquids (helium and hydrogen) have the great advantage that they can be made extremely pure (especially helium). However, it is still necessary to use some special method to eliminate the possibility of nucleation on the walls.

The normal freezing temperature of hydrogen is $T_f = 13.9$ K. By extrapolating the measured specific heat of liquid hydrogen to lower temperatures we estimated that the difference in energy between liquid and solid hydrogen at $T = 0$ K was only 6.5 K [6]. This is very small compared to the 90 K total binding energy of the solid [7] and suggested that it might be possible to supercool liquid hydrogen far below T_f. We then calculated the rate at which nucleation of the solid phase should occur in supercooled liquid. In the region just below T_f the energy barrier $E_{barrier}$ decreases rapidly as T goes down giving a rapid increase in $\Gamma_{thermal}$. However, far below T_f the barrier becomes constant and then, since the energy kT available to overcome the barrier is decreasing, the nucleation rate due to thermal fluctuations becomes very small. The nucleation rate then falls to a low temperature value Γ_0 which is determined by the rate of quantum tunnelling through the nucleation barrier.

This dependence of the nucleation rate on temperature raised the intriguing possibility that if hydrogen could be cooled quickly through the temperature range in which the nucleation rate was high (roughly 4 to 8 K), then perhaps the liquid could be cooled down to arbitrarily low temperatures. To give the best chance of this working it would be best to use a very small volume of liquid and cool it quickly through the temperature range in which the nucleation rate was large. It was also necessary to eliminate nucleation on the container walls. To achieve this we inserted drops of liquid hydrogen into helium fluid, i.e., helium above the critical point of 5.2 K [8]. The helium was much more compressible than the hydrogen and so by adjusting the pressure applied to the helium we could make the densities match and have the hydrogen drops float in the helium far away from the walls. The smallest drops that we could study had a radius of 50 µ and these could be cooled to only about 10.7 K before solid nucleated. At this temperature the nucleation rate was $\sim 10^7$ cm^{-3}s^{-1}. It was difficult to use smaller drops or cool faster using this approach.

A second approach was to first magnetically levitate a large drop of liquid helium (of the order of 1 cm^3) at a temperature of 1K, for example, and then to condense hydrogen molecules onto this drop. In this way, bulk hydrogen liquid would be first formed at low temperatures and would not have to be cooled through the temperature range where the nucleation rate was high. For various reasons we never completed this experiment (we still hope to!), but the helium levitation experiment did lead to many interesting results [9]. One remarkable result was that under certain conditions two levitated superfluid helium drops would approach and appear to make contact but not coalesce. A photo of this is shown in Fig. 1. On occasion two drops would even bounce off of each other. The detailed explanation of why coalescence did not occur is given in ref. [10]

Since this work there have been many attempts to make low temperature liquid hydrogen using much smaller helium droplets formed from a jet rather than magnetically levitated. For a review of the status of this field, see ref. [11].

Figure 1. Two levitated helium drops touching each other. The drops are viewed from above. The drop on the right is attached to the capillary used to introduce liquid into the cell.
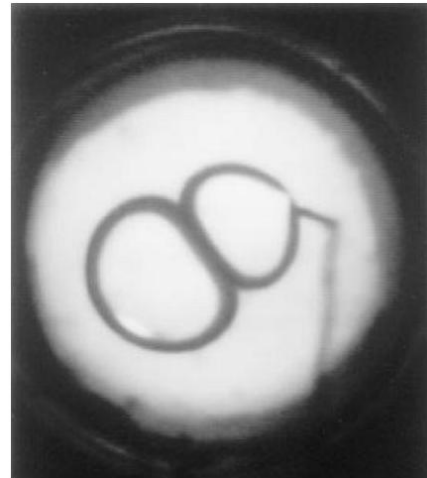
## II. Liquid helium at negative pressures

We then began to think about what liquid helium might be like under negative pressure. There had been many reports of studies of the formation of bubbles in liquid helium [12]. In most of these experiments a sound pulse was applied to the liquid giving positive and negative pressure swings. The formation of bubbles on the negative swing was monitored either optically or by detecting the acoustic noise produced when the bubbles collapsed. In these experiments it had been found that bubbles first appeared at a threshold negative pressure that was just a small fraction of 1 bar. However, these experiments had been performed using helium in the main helium bath of a glass dewar, and mostly with the sound applied to a large volume of the liquid. Under these conditions, it was certainly possible that impurities, such as small particles of solid air, could provide sites for heterogeneous nucleation. In 1989, Nissen *et al.*[13] published a paper describing measurements in which a hemispherical ultrasonic transducer was used to generate sound that came to a focus within a volume of only $\sim 10^{-5}$ cm^3. This small volume reduced the chance that impurities can affect the measurement. They found that bubbles were first formed at a negative pressure which became larger as the temperature decreased. They estimated that the pressure reached a value of about -8.1 bars at 1.6 K.

We had already made some rough theoretical estimates of the properties of helium at negative pressure and we were surprised by the large magnitude of this negative pressure. When a negative pressure is applied to a liquid, the bulk modulus B and the sound velocity c both decrease. At a critical negative pressure P_s (the spinodal pressure), B and c go to zero and the liquid becomes unstable against long wavelength density fluctuations. One can try to estimate the value of P_s by extrapolating the measured values of c for positive pressure into the negative pressure range. If the energy of the liquid can be represented by a polynomial in the density so that close to the spinodal density ρ_s the pressure varies as $P - P_s \propto \left(\rho - \rho_s \right)^2 + ..$, then it is easy to show that for pressures slightly greater than P_c the sound velocity should vary with pressure according to

$$c^4 \propto P - P_s \qquad (4)$$

By fitting experimental data to this law we obtained a value for P_s of around -9 bars at $T = 0$ K [14,15]. Monte-Carlo calculations [16,17] and density-functional calculations [18] give values which are consistent with this value of the spinodal pressure.

To develop a more detailed theory of nucleation we constructed a density-functional theory [14] to describe not only the properties of bulk uniform helium, but also non-uniform liquid; this is required in order to estimate the nucleation barrier and nucleation rate. More accurate density-functional calculations were performed by the group of M. Barranco in Barcelona [19]. The calculations were later extended to include nucleation by quantum tunnelling [20] and the possible effects of quantized
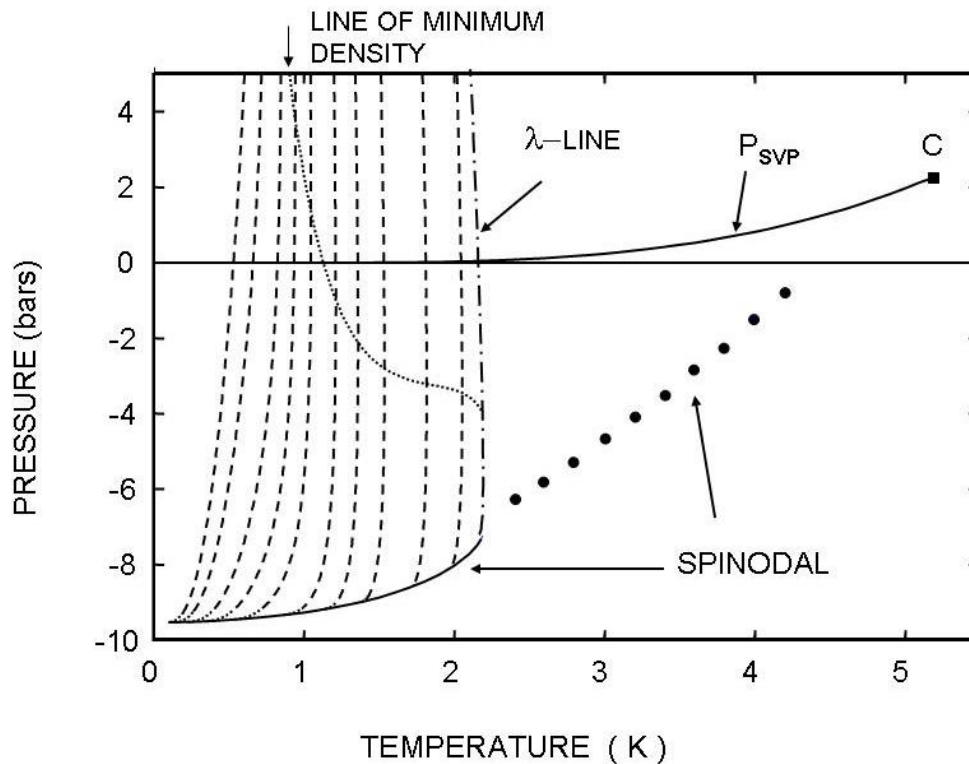
Figure 2. The calculated phase diagram of helium as estimated in ref. [25]. The dashed lines are lines of constant entropy for entropies of 0.001, 0.002, 0.005, 0.01, 0.02, 0.05 0.1, 0.2 0.5, 1.0 and 2 J g^{-1} K^{-1}. The spinodal for the normal liquid (solid circles) is from the estimate made in Ref. [15].The curve showing the saturated vapor pressure P_{SVP} and the location of the critical point are the standard experimental values.

vortices on nucleation [21]. These calculations predicted that because of the effect of the spinodal, nucleation should occur at negative pressures of somewhat smaller magnitude than had been found by Nissen *et al.* [13]. A review of more recent measurements of the cavitation threshold has been given by Balibar [22]. The results are in reasonable agreement with theory although the accurate estimation of the negative pressure that is reached in these ultrasonic experiments remains a challenge [23]. The measurements confirm the estimates of the spinodal pressure in both helium-3 and helium-4 [23] and are consistent with the assumption that nucleation near $T = 0$ K takes place as a result of quantum tunnelling [24].

With D.O. Edwards, I then made a calculation of the phase diagram of helium-4 as a function of temperature throughout the negative pressure regime [25]. The result is shown in Fig. 2. This calculation involved putting together many pieces including an estimate of the roton-phonon dispersion curve, the effect of interactions between rotons as the lambda transition was approached, etc. While all of the details are described in the original publication, there are several points of special interest to mention.

1) It turns out that the sound velocity as a function of pressure is rather poorly described by Eq. 4, but is fit with remarkable accuracy by the law

$$c^3 \propto P - P_s \qquad (5)$$

This is shown in Fig. 3. Equation 5 implies that for a density slightly greater than the spinodal density the pressure varies with density as

$$P - P_s \propto \left(\rho - \rho_s \right)^3 + .. \qquad (6)$$

It was possible to understand this behaviour based on a renormalization group theory in which the contribution of zero-point fluctuations to the internal energy of the liquid was treated in a self-consistent way. But although this is a theory for <u>near</u> the spinodal it is found that the law of Eq. 5
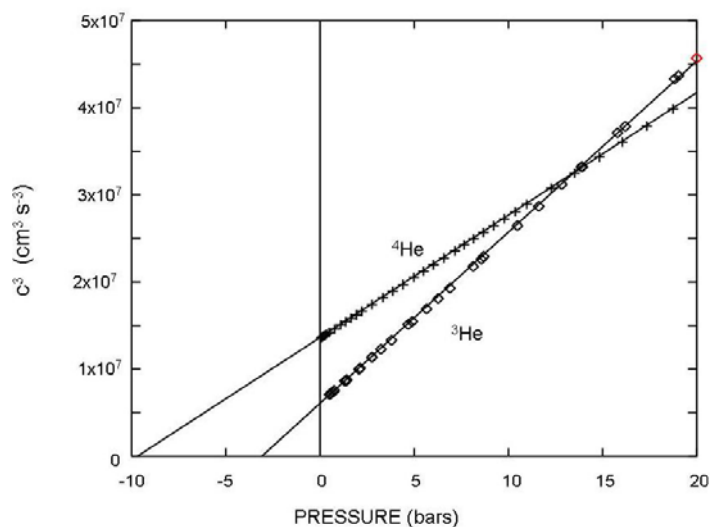
Figure 3. Plots of the cube of the sound velocity as a function of pressure for helium-3 and for helium-4.
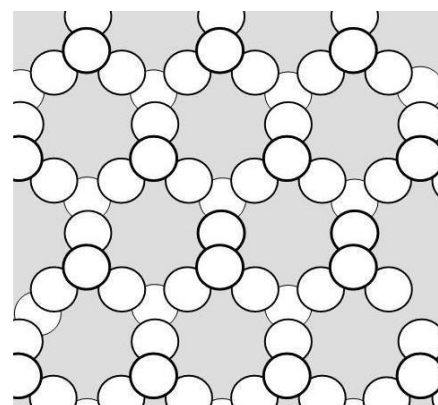
Figure 4. The T_d^2 structure proposed by Keesom and Taconis as a model for the arrangement of atoms in liquid helium.

holds with great accuracy up to the freezing pressure for both liquid helium-3 and for helium-4. It even holds remarkably well for solid helium-4 up to a pressure of 150 bars! The law seems to be a very general feature of highly quantum systems. But to date there is no explanation of this.

2) For most crystalline solids the phonon dispersion relation is linear for very small momentum and then bends over, i.e., the first correction to the dispersion is negative. However, as already mentioned for helium at zero pressure the dispersion relation curves slightly upwards but curves down at higher pressures. Once the existence of the spinodal was recognized it became easier to understand. Near to the spinodal the sound velocity goes to zero and so certainly the dispersion relation must curve upwards. Thus, one can consider that the positive dispersion at zero pressure comes about because, in some sense, liquid helium at zero pressure is near to the spinodal.

3) In his classic book on superfluids [26], F. London discussed the arrangement of atoms in liquid helium. His goal was to understand how helium can "hold together" considering its large molar volume. He notes that if helium at $P = 0$ had a close packed structure the spacing between nearest neighbour atoms would be $a = 2^{1/6} (V / N)^{1/3} = 4.0$ Å. However, when the spacing between two helium atoms exceeds 3.2 Å the effective spring constant between atoms is negative and so, at least based on classical mechanics, such a structure would be unstable. He then discusses the idea that helium has a much more open structure in which each atom has only 6 nearest neighbours; this had been proposed earlier by Keesom and Taconis [27] and is shown in Fig. 4. With this structure the interatomic spacing is 3.16 Å. London emphasized that he is not implying that the liquid is crystalline; he is simply giving an example of an atomic configuration that has the same volume per atom as the real liquid. But now consider helium close to the spinodal. For helium-4 the molar volume at the spinodal is 42.5 cm³ and for helium-3 it is 56 cm³! How does the liquid withstand a negative pressure at these molar volumes? It would be of great interest to investigate, for example, by computer simulation, how the average coordination number and the atomic arrangement changes as the spinodal is approached. Some preliminary results on this have been obtained by Boronat [28].

4) Given that there may be a significant change in the structure of the liquid near to the spinodal, are there some new phases that appear in this pressure range? For example, in the case of helium-3 the transition temperature to the superfluid B-phase decreases as the pressure goes down. Based on a simple extrapolation of the transition temperature, it appears that this temperature goes to zero at a critical pressure P_B which is around -3 bars. If P_B is in fact positive with respect to P_s (-3.14 bars)

then decreasing the pressure at zero temperature results in a quantum phase transition. But it is also possible that for some reason P_B and P_s are equal.

How can one test these theoretical ideas? The most detailed tests performed so far come from the measurements of the negative pressure at which nucleation occurs. The analysis of these measurements is complicated because it is necessary to estimate the pressure that is reached and also to allow for the change in temperature that occurs when the liquid is expanded. Nevertheless, overall these experiments have given results in good agreement with experiment and support the estimated value of the spinodal pressure.

III. Electrons and negative pressures

The use of negative pressures has opened up completely new possibilities for the study of electrons in liquid helium as we will now describe. It was Richard Ferrell who in 1957 [29] first proposed that an electron in helium may exist in a "bubble state". Because an electron would prefer to be in vacuum rather than in helium (energy difference 1 eV), it is energetically favorable for an electron in helium to force open a cavity in the liquid which is free of helium atoms. As a first approximation, one can consider that the energy of the electron bubble is given by the expression

$$E = E_{el} + \alpha_{lv} A + PV \tag{7}$$

where E_{el} is the energy of the electron ($h^2 / 8mR^2$ for an electron in the 1S ground state of a spherical bubble), A is the surface area of the bubble, V is the volume and P is the pressure. For the 1S state at zero pressure the bubble is in a state of mechanical equilibrium when the radius is $R_{eq} = 19$ Å. A more accurate theory of the bubble can be constructed using density-functional theory [30,31,32].

These bubbles have been studied in many experiments. The size can be checked through a measurement of the photon energies required to excite the electron to higher energy quantum states (1P and 2P) [33,34,35,36]. For a moving bubble the phonons and rotons in the liquid exert a drag force which is proportional to the radius squared. Mobility measurements therefore provide information about the bubble size and give results which are in good agreement with theory [37,38,39]. If a positive pressure is applied an electron bubble becomes smaller. For a negative pressure the bubble grows and at a critical negative pressure given by

$$P_c = -\frac{16}{5}\left(\frac{2\pi m}{5h^2}\right)^{1/4} \alpha^{5/4} \approx -1.9 \text{ bars,} \tag{8}$$

the bubble becomes unstable and begins to grow without limit. This result was first derived by Akulichev and Bulanov [40]. We have been able to detect these electron "explosions" and confirm that they do indeed occur at the expected negative pressure [30]. In the first experiment along these lines a hemispherical ultrasonic transducer was used as already described. In this situation an electron explosion will be seen only if an electron happens to be in the small region around the acoustic focus. The period of the sound is in the range around 1 µs and so the pressure is changing on a time scale which is very long compared to the time scale for vibrations of the bubble ($\sim 10^{-11}$ s). Thus, the explosion of the bubble is essentially a quasi-static process.

In more recent experiments [41] we have used a sound transducer to generate a planar sound pulse which as it passes through a volume of several cubic centimeters explodes every electron bubble in its path. By applying a series of sound pulses (the rate in the first experiment was 4 per second), a movie can be made showing the motion of individual electrons. Two frames from one such movie are shown in Fig. 5. In these pictures the sound transducer is at the top and the electrons are always seen to move down the cell.
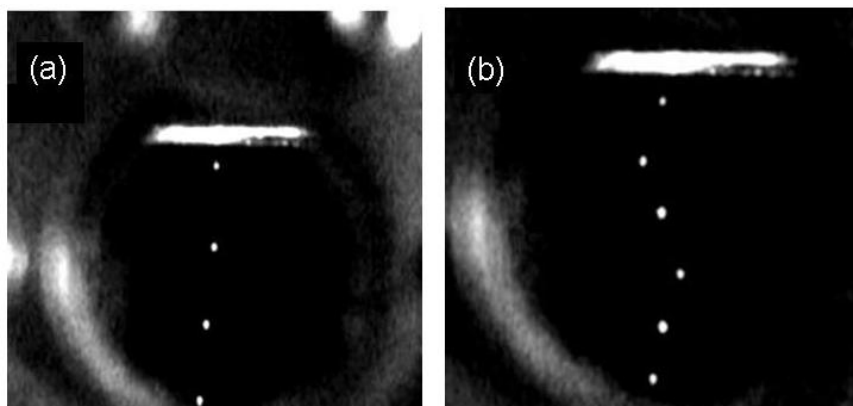
Figure 5. Two frames from a movie of an electron moving through superfluid helium. In part a) the electron is being dragged down the cell as a result of the flow of the normal fluid. In b) the motion of the electron is modified by trapping on a quantized vortex.
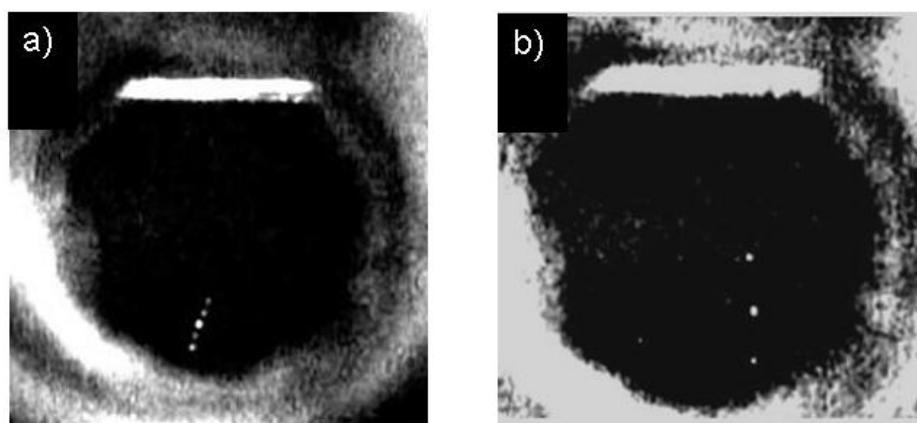
Figure 6. Electrons that are first seen in the interior of the liquid helium cell and which are the result of Compton scattering of the background gamma radiation in the laboratory.

In this experiment the experimental cell contained no source of electrons and so it was interesting to investigate where the electrons came from. At first sight, it was surprising that the majority of the electron tracks began at the bottom surface of the sound transducer at the top of the cell. We were able to show that the electrons were mostly produced by an interesting mechanism involving cosmic rays. A cosmic ray muon passing through the liquid causes a large amount of ionization along its track. However, essentially all of these ions recombine and thus no free electrons are left in the liquid. But when recombination occurs, high energy photons are produced which can travel without attenuation through the liquid and lead to photoemission at the transducer surface. These are the electrons that we see. They move down the cell because when the sound transducer is driven energy is dissipated resulting in flow of normal fluid down the cell which drags the electron bubbles with it.

While most of the electrons that are seen follow slightly curved paths corresponding to the flow of the normal fluid, a small fraction have snake-like paths as shown in Fig. 5b. It appears that these electrons are attached to quantized vortices; the observed motion is presumed to be the sum of the motion of the electron along the vortex line together with the motion of the vortex itself.

A small number of electrons are first detected far from the walls of the cell (see Fig. 6). These electrons appear to be the result of Compton scattering of gamma rays coming from outside the

cryostat. We have confirmed this by bringing a gamma source up to the outside of the cryostat and thereby greatly increasing the rate of such electrons.

Now we mention some of the unsettled questions in this field; for a more detailed discussion see ref. [42]. One question arose in the first experiment in which electron bubbles were exploded using a focusing transducer. A small number of explosions were observed even when the sound amplitude was not large enough to reach the critical pressure of Eq. 8. it was determined that these explosions came about as follows [30]. In the experiment a β-source was used to inject electrons into the helium. When an electron came to rest in the liquid, it would force open a cavity which would grow rapidly, reach a volume larger than the equilibrium size, and then relax back to have the equilibrium radius R_{eq}. Thus for a few picoseconds the electron bubble will have a radius larger than R_{eq} and can be exploded by a pressure of smaller magnitude than P_c.

A second question concerns the interaction of electrons with vortices. It is well known that at low temperatures, electrons in helium can become attached to quantized vortices [43]. It is possible to measure the pressure P_c^{vort} at which these electrons explode and compare it with the explosion pressure P_c (see Eq. 8) for electrons in the bulk of the liquid [30]. It is found that P_c^{vort} is about 13 % less than P_c whereas theory gives a difference of only 5%[30,44]. This difference is disturbing because an understanding of the interaction of foreign particles with vortices is currently of great interest for the interpretation of many experimental studies of the hydrodynamics of liquid helium [45]. In addition to this problem, it is found that below 1 K there appear to be some electron bubbles which are easier to break even than the electrons attached to vortices [46]. There is currently no understanding of the nature of these objects; possibly they are bubbles attached to more than one vortex, but to date this has not been demonstrated.

A third question concerns the so-called exotic ions. Time-of-flight mobility measurements performed by Ihas and Sanders [47] and others [48] have revealed that in addition to the normal electron bubble there are a large number of other negatively charged objects (at least 12) with a radius between about 8 and 16 Å. These ions have been seen when the source of electrons has been an electrical discharge above the surface of the liquid. These objects have discrete mobilities; this indicates that they have discrete sizes, not a continuous distribution. At the present time there is no understanding of the physical nature of these objects. One possible way to determine their structure may be to apply a negative pressure and find out the pressure at which each ion explodes.

A final mystery concerns how electron bubbles are affected by light. As already mentioned, it is possible to excite the electron to a higher energy state, such as 1P or 2P. When this is done, the outward pressure exerted by the electron on the helium changes and the bubble shape is modified. Because the wave function no longer has spherical symmetry, the bubble loses spherical symmetry. It is straightforward to calculate this shape, and results for the 1P state at different pressures are shown in Fig. 7 [49]. Note that the bubble has a distinct waist because the wave function of the electron vanishes in the $z = 0$ plane. These objects are referred to as peanut bubbles. Because the electron is now pushing outward harder than in the 1S state, the 1P bubbles explode at a smaller negative pressure; this is calculated to be -1.63 bars [50]. We have been able to excite to the 1P state (the CO_2 laser has the right wavelength to do this) and confirm that this does indeed produce 1P bubbles which explode at the predicted decreased negative pressure [51]. When these experiments are performed in the temperature range from about 1.5 K up to near the lambda point, the liquid contains many thermal excitations (phonons and rotons) and thus the damping of the motion of the bubble wall is large. As a result after the light is absorbed the bubble shape relaxes smoothly towards the new equilibrium shape. However, at lower temperatures the damping becomes small and the bubble overshoots the equilibrium shape. If the pressure is larger than about 1 bar, theory then predicts that the bubble will break into two "baby bubbles", and it is not known what happens next. In our experiments, we have confirmed that under these conditions, i.e., $T < 1.5$ K and pressure > 1 bar, optical excitation to the 1P state does not result in the production of any peanut bubbles; this confirms the idea that bubble
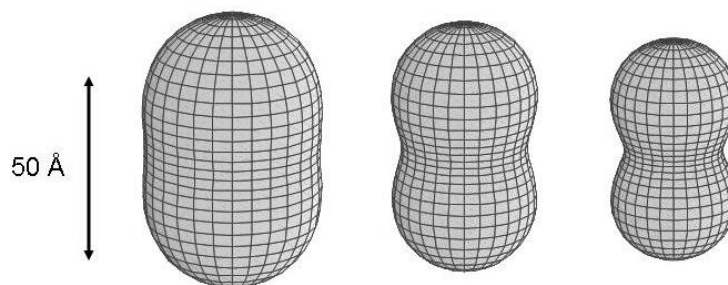
Figure 7. The shape of 1P electron bubbles at pressures of -1, 2 and 5 bars.

fission takes place, but does not determine what is produced. Does part of the wave function of the electron become trapped in each baby bubble? [52]If it does, what happens next? Does the helium make a measurement, find the electron in one bubble, and cause the wave function in the other bubble to collapse? And, if this does indeed happen, at what time does it occur? Simulations in which collapse is included show that the motion of the surviving bubble <u>after</u> the collapse is affected by the time at which the collapse occurs and so it may be possible to determine this time experimentally [53].

Acknowledgments

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