

# Looking back at superfluid helium

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A few years after the discovery of Bose Einstein condensation in several gases, it is interesting to look back at some properties of superfluid helium. After a short historical review, I comment shortly on boiling and evaporation, then on the role of rotons and vortices in the existence of a critical velocity in superfluid helium. I finally discuss the existence of a condensate in a liquid with strong interactions, and the pressure variation of its superfluid transition temperature.

## The discovery of superfluidity

In January 1938, J.F. Allen and A.D. Misener [1] published the experimental evidence that the hydrodynamics of liquid helium was not classical below 2.2 K. In the same issue of *Nature*, Kapitza introduced the word “superfluid” to qualify this anomalous behavior [2]. Soon after this discovery, F. London suggested that superfluid helium forms a macroscopic liquid matter wave, as a consequence of “Bose-Einstein condensation” (BEC). 65 years later, it is generally accepted that London was right, but it happened to be very difficult to prove. On the contrary, when BEC was discovered in cold alkali vapors (in 1995), the experimental evidence was immediately very clear. The superfluidity of these cold gases was soon demonstrated as well. It is thus interesting to look back at some properties of superfluid helium now. As we shall see, not everything is completely understood in the properties of superfluid helium. In this short review, I have selected only a few properties of superfluid helium. For a complete description, one can look at the historical book written by J. Wilks [3], or the one by P. Nozières and D. Pines on Bose liquids [4]. Here, I only wish to recall some aspects of the early history of this subject in order to show that fundamental questions came up very soon in this field. Then, I wish to briefly discuss three particular topics:

1- boiling and evaporation because this is what makes superfluid helium look different from normal helium in a dewar.

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2- the mechanisms which determine the “critical velocity”

3- the existence of a condensate in superfluid helium and the pressure variation of the superfluid transition temperature.

In the years 1928-32 in Leiden, W.H. Keesom and his co-workers had already realized that liquid helium exhibited surprising properties below about 2.2 K [5]. They had found a peak in the temperature variation of the specific heat at this temperature. The shape of this peak resembling the greek letter lambda, Keesom called the superfluid transition temperature the lambda point or “lambda temperature  $T_\lambda$ ”. He also called “helium I” the liquid above  $T_\lambda$  and “helium II” the liquid below  $T_\lambda$ . In his laboratory in Leiden, Keesom had also discovered that helium II was able to flow through very tiny pores as soon as in 1930 [6]. In 1932 in Toronto, J.C. McLennan, H.D. Smith and J.O. Wilhelm then discovered that liquid helium ceased boiling when cooled down through  $T_\lambda$  [7]. This was soon attributed to an exceptionally high thermal conductivity, as evidenced by successive experiments by W.H. Keesom again [8] and by J.F. Allen in Cambridge [9]. J.F. Allen had been hired with R. Peierls to replace Kapitza there, because Kapitza had been forced by Stalin in 1934 to stay in Moscow and to quit his research position in Cambridge[10]. Since the high thermal conductivity was attributed to some kind of convection or turbulence, researchers looked at the flow of liquid helium. In 1935 in Toronto, J.O. Wilhelm, A.D. Misener and A.R. Clark had measured the viscosity of liquid helium with a torsion pendulum and found that it decreased sharply below  $T_\lambda$  [11]. When A.D Misener moved to Cambridge in order to prepare his doctorate with J.F. Allen, they started together a systematic study of the flow of liquid helium through capillaries. Classical fluids obey the Poiseuille law: the flow rate is proportional to the pressure difference across the capillary and to the fourth power of the capillary radius. Instead of this, Allen and Misener found that, below  $T_\lambda$ , the flow rate was not only high, it was independent of the pressure and independent of the capillary radius which they had changed by a factor 50! Clearly, this liquid was not classical. In a sense, this was also what Kapitza claimed in his article [2], since he introduced the word “superfluid” in connection with the already known “superconductivity”. However, Kapitza did not justify his remarkable intuition that superfluidity and superconductivity were related phenomena, and his 1938 article contained no quantitative measurements. He performed remarkable experimental measurements during the following years.

In february 1938, J.F. Allen and H. Jones published another astonishing discovery. They had found a remarkable thermomechanical effect [12]. When heating superfluid helium on one side of a porous medium or a thin capillary, the pressure increased sufficiently to produce a little fountain at the end of the tube which contained the liquid.

The “fountain effect” was another spectacular phenomenon which was impossible to understand within classical thermodynamics. This is really what triggered Fritz London’s thinking [13]. London new that, in this liquid, there were large quantum fluctuations responsible for the large molar volume, and he proposed that the superfluid transition at  $T_\lambda$  was a consequence of Bose-Einstein condensation (BEC). 65 years later, the connection between superfluidity and BEC is still a matter of debate and study. Before discussing this (in Section III), I wish to say a few words about boiling and evaporation (section I), and , in Section II, I briefly review the problem of the critical velocity, in connection with Landau’s “two fluid model” [14], the existence of “rotons” and quantized vortices. One of my motivations in section II is to allow a comparison with observations of a critical velocity and vortices in gaseous superfluids.

## BOILING, EVAPORATION AND CAVITATION

The easiest way to see superfluidity is to look at liquid helium in a dewar, while pumping on it so as to cool it below  $T_\lambda = 2.2$  K (see Fig. 1). When crossing  $T_\lambda$ , the liquid stops boiling. This is because the thermal conductivity of liquid helium has suddenly increased, so that the temperature is very homogeneous. The walls are no longer warmer than the surface. They no longer provide efficient nucleation sites for bubbles. As a result, superfluid helium evaporates from its free surface, instead of showing bubble nucleation on hot defects. Superfluid helium looks quiet, and it is tempting, although not rigorous, to take this as an illustration of quantum order getting established. At least this shows a striking reality: a liquid made of very simple atoms can exist in two different states! It happens that I worked on the evaporation of superfluid helium [15] so that i would like to add a few comments on the evaporation of superfluid helium, an interesting property which is rarely mentionned. We have verified the prediction by P.W. Anderson that, at low enough temperature, this evaporation is a quantum process which is analogous to the photoelectric effect [16]. A photon incident on a metal surface can eject one electron elastically: the electron kinetic energy is the difference between the photon energy and the binding energy of the electron to the metal. Similarly, we have shown that atoms can be ejected from superfluid helium with a kinetic energy which is close to 1.5 K, the difference between the energy of incident rotons (whose minimum is 8.65 K) and the binding energy of atoms to the liquid, which is nothing but the latent heat of evaporation per atom (7.15 K). This phenomenon was later called “quantum evaporation” by A.F.G. Wyatt who made a long quantitative study of it. A few open issues remain in this subject, such as the exact measurement and calculation of evaporation probabilities by phonons or rotons as a function of energy, momentum

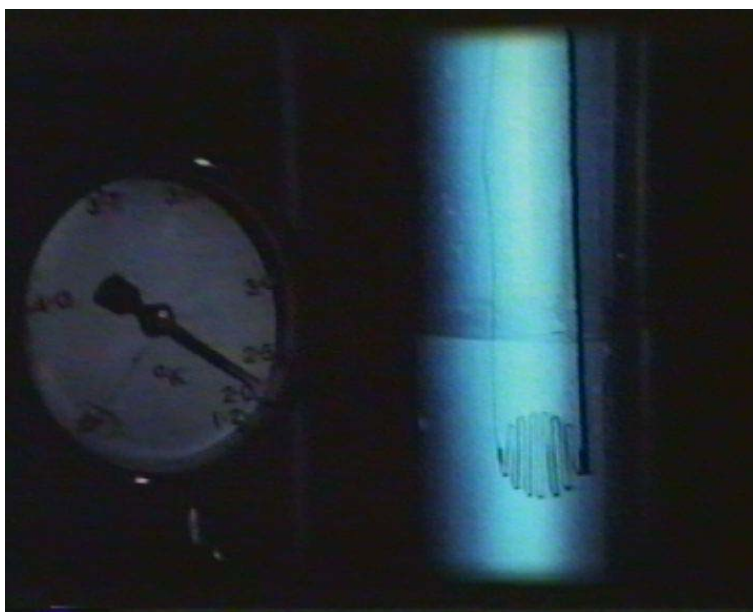
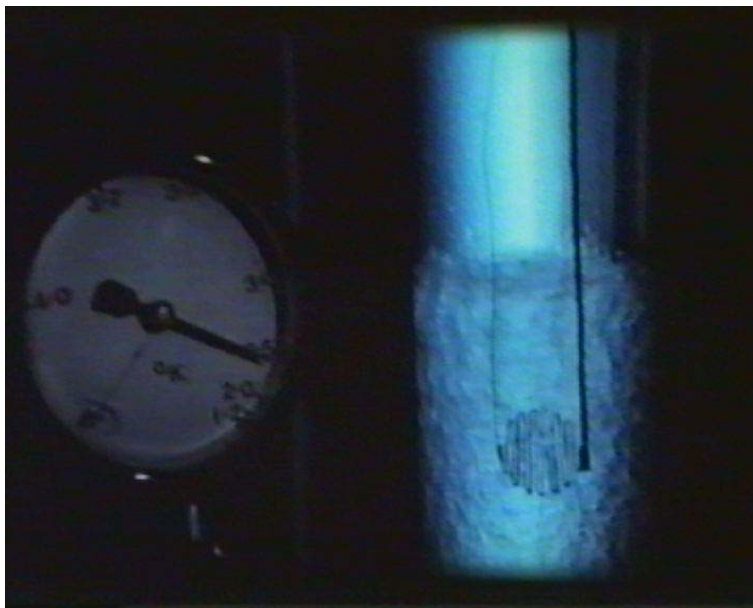


FIG. 1: As shown by these two images from a film by J.F. Allen and J. Armitage, superfluid helium stops boiling below  $T_\lambda$ . This is due to its large thermal conductivity. The top image is taken at 2.4 K as indicated by the needle of the thermometer on the left. The bottom image is taken just below the lambda transition.

and incidence angle, or the nature of possible inelastic processes as well.

Furthermore, one should not believe that boiling, more generally cavitation or bubble nucleation, is impossible in superfluid helium. On the contrary, this is another interesting subject on which we also worked [34]. A strong local

heat input can generate bubbles in superfluid helium, of course. Moreover, one can apply a large stress, that is a large negative pressure to liquid helium at low temperature and also look for the nucleation of bubbles. This is a way to test the internal cohesion of this liquid. We have observed this phenomenon near -9.5 bar, the stability limit of liquid helium under stress, also called the “liquid-gas spinodal limit”. It allowed us also to obtain some information on superfluidity at reduced density as we shall see briefly in Section III.

### THE CRITICAL VELOCITY: ROTONS AND VORTICES

The vanishing of boiling is spectacular but the existence of flow without dissipation is of course the more fundamental property which signals superfluidity. In may 1938, one month only after London had published his suggestion of a Bose-Einstein condensation, L. Tisza introduced a “two fluid model” which qualitatively explained the observations. He considered that “the atoms belonging to the lowest energy state do not take part in the dissipation. Thus the viscosity of the system is entirely due to the atoms in the excited states”. He understood that this liquid could be considered as a mixture of two components: a superfluid component which carries no entropy and has zero viscosity, plus a normal component, made of atoms in excited states, which is viscous and conducts heat. The ratio of one to the other component was determined by the temperature: at zero temperature there is only the superfluid and at the lambda transition there is only the normal component. The important point was to understand that these two components could move independently. This very pure liquid had two independent velocity fields, something London could not easily admit. From these considerations Tisza predicted that when superfluid helium flowed through a capillary, the normal component, being viscous, was blocked while the superfluid component could flow without dissipation. As a consequence, viscosity measurements could not give the same result when done in an open geometry, as in the torsion pendulum used in Toronto, or in a restricted geometry like a thin capillary or a porous medium. This resolved the apparent discrepancy between different experiments at that time. Moreover, he made an even more surprising prediction, namely that “a temperature gradient should arise during the flow of helium II through a thin capillary”. Tisza was right! Helium cools down when flowing so. Furthermore, he explained the fountain effect as the reverse of the previous one: the superfluid component moves in the direction of the hot side where the pressure increases. His 1938 article was only one column long. He developed his “two fluid model” in two subsequent articles [18] where he made another fundamental prediction: thermal waves should exist where the superfluid and normal components move out of phase.

Nearly all the predictions by Tisza were proved correct by subsequent experiments. However, his model appeared slightly incorrect because the superfluid and normal components could not be defined in relation to a Bose gas. The two fluid model was subsequently developed by Landau in his 1941 article, with no reference, this time, to BEC. He considered energy states of the liquid instead of individual atoms. He attributed a mass density  $\rho_s$  to the part of the fluid which occupies the ground state. The rest of the density  $\rho_n = \rho - \rho_s$  corresponded to the excited states. Furthermore, he had a remarkable intuition about the nature of these excited states. He assumed that single particle states were replaced by collective modes of two different kinds: phonons, i.e. sound quanta, and quantized vortices which he called “rotons”. Phonons had basically a linear dispersion relation  $\omega = ck$  and rotons had a quadratic one with a gap ( $\epsilon = \Delta + p^2/2\mu$ ), so that no excitation had a phase velocity  $\epsilon/p$  less than a certain minimum value  $v_c$ . From this assumption, Landau calculated the thermodynamics of helium II, the magnitude of thermal waves called “second sound”, etc. The major breakthrough in this article is the introduction of elementary excitations as quantized collective modes which led him to the crucial prediction of a critical velocity  $v_c$ . He explained that, due to the necessary conservation of energy and momentum, the superfluid component in motion at a velocity  $v$  could not slow down by exciting collective modes if  $v$  was less than  $v_c$ . In the absence of rotons,  $v_c$  would have been the sound velocity  $c$ . Landau modified the dispersion relation of rotons in his 1947 article [19] in order to obtain a better agreement with thermodynamic properties. The phonon branch evolved continuously into a roton branch  $\epsilon = \Delta + (p - p_0)^2/2\mu$  around the finite momentum  $p_0$ . Given this phonon-roton spectrum, Landau’s critical velocity was predicted to be about 60 m/s at low pressure.

M. Cohen and R.P. Feynman [20] then proposed to verify Landau’s dispersion relation in neutron scattering experiments. This was done with great accuracy by several groups, in particular by Henshaw and Woods [21]. Of course, it was a strong support to Landau’s theory. In the mean time, superfluidity was proved not to exist in liquid helium 3 at similar temperatures, and this was a strong support to London and Tisza. As for the existence of Landau’s critical velocity, things appeared difficult. In fact, Landau had proposed this mechanism but he had not ruled out the possibility that other mechanisms could exist at lower velocity. The existence of quantized vortex lines was proposed in 1949 by Onsager [22] who predicted that the circulation of their velocity should be quantized as multiples of  $h/m$ . Independently in 1955, Feynman [23] explained that such vortices should lead to a much lower critical velocity of order  $v_c = (\hbar/md) \ln(d/a)$  where  $d$  is the size of the container (a vessel or a capillary diameter) and  $a$  the atomic size of the vortex core. Feynman was right. The most beautiful evidence for the vortex mechanism was obtained by

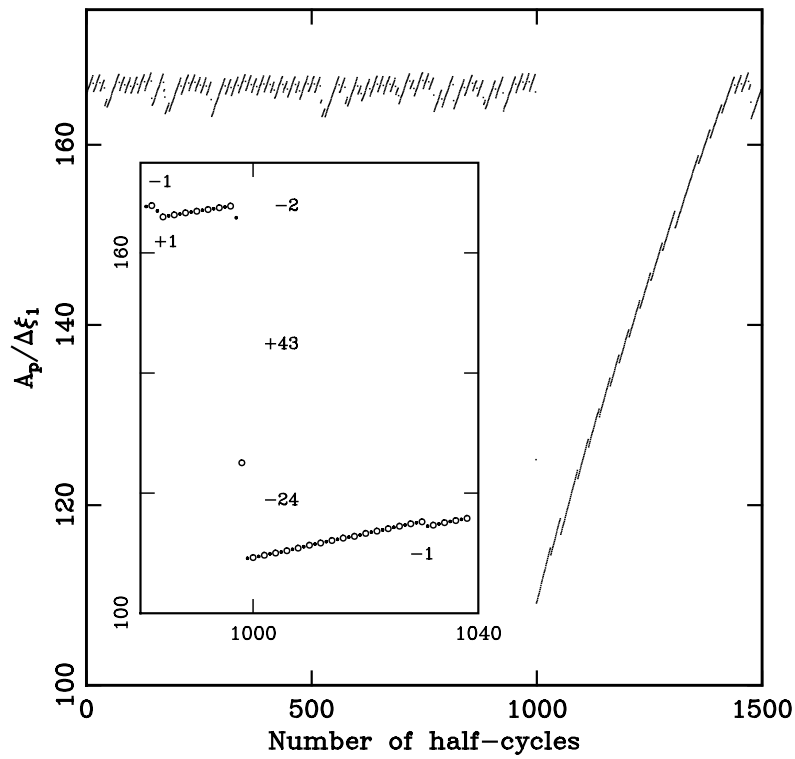


FIG. 2: The quantized dissipation in the experiment by O. Avenel and E. Varoquaux. The vertical coordinate is the amplitude of oscillation of a superfluid flow through a small aperture. It is proportional to an average velocity in the hole. The horizontal coordinate is proportional to time. The velocity changes by finite jumps which are quantized. Single jumps correspond to one quantized vortex line crossing the hole. Multiple jumps are also observed.

Avenel and Varoquaux [24] who could see quantized dissipation events in a flow through a small aperture (see Fig. 2). Moreover, their analysis of vortex nucleation shows a remarkable crossover from thermally activated nucleation above a crossover temperature of order 0.2 K to nucleation by quantum tunneling in the low temperature limit. A similar crossover was found in studies of superconducting Josephson junctions, also in our studies of cavitation in superfluid helium 4.

As for Landau's critical velocity, it was also observed, but in cases where the system size was very small only. P. McClintock et al. measured the mobility of electrons moving in superfluid helium under special conditions. These electrons repel the electronic clouds of neighbouring atoms so as to form little empty cavities around them, whose typical radius is  $10 \text{ \AA}$ . At low pressure and small electric field, these electron bubbles trap vortex rings as was checked from by their energy vs momentum curve. At high pressure (25 bar, near the melting pressure) and high electric field (from 20 to 2000 V/cm), the electron bubbles could in fact be accelerated to velocities slightly larger than Landau's prediction. This was understood by Bowley and Sheard as a consequence of rotons being emitted in pairs [25]. This

whole subject would need a much longer review, but the comparison with gaseous superfluids is interesting. Superfluid gases have no rotons in their excitations. Landau's critical velocity is thus associated with phonons. It is observed if the moving object is small (an atom), but if it is larger (a laser beam for example), then dissipation occurs at lower velocity. Just as in superfluid helium the nucleation of vortices is invoked to understand the low value of the critical velocity in this case. The comparison between quantized vortices in superfluid helium and in superfluid gases has been beautifully extended to the observation of similar vortex arrays in both cases (see J. Dalibard and C. Salomon, this conference).

### THE CRITICAL TEMPERATURE AND THE CONDENSATE FRACTION IN LIQUID HELIUM

London had noticed that  $T_\lambda$  was close to the transition temperature of an ideal Bose gas with the same density.

$$T_{BEC} = \left( \frac{2\pi\hbar^2}{1.897mk_B} \right) n^{2/3} \quad (1)$$

Inserting a number density  $n = 2.18 \times 10^{22} \text{ cm}^{-3}$  in Eq. 1 leads to  $T_{BEC} = 3.1 \text{ K}$  while  $T_\lambda = 2.2 \text{ K}$ . London had also noticed that the molar volume of liquid helium was large, due to the large kinetic energy corresponding to the quantum fluctuations of atoms in the cage formed by their neighbours. He had further noticed that the anomaly of the specific heat of liquid helium near  $T_\lambda$  was not very different from the cusp one should expect in the case of the BEC of an ideal gas. He thus claimed that, for the understanding of the superfluid transition in helium, "it seems difficult not to imagine a connection with the condensation of the Bose-Einstein statistics". However, in the next sentence, he immediately added that "a model which is so far away from reality that it simplifies liquid helium to an ideal gas" cannot lead to the right value of its specific heat.

In fact, the difficulty is much deeper than a question of specific heat only. As claimed by A.J. Leggett [26], it is not rigorously proven that a condensate, i.e. a macroscopic occupation of a ground state, should necessarily exist in a Bose liquid. It is not obvious either that there is a continuous path from the ideal Bose gas to the strongly interacting Bose liquid via the weakly interacting Bose gas where the existence of a condensation has been demonstrated both theoretically and, now, experimentally. The total absence of reference to BEC in Landau's work is obviously intentional. Landau deliberately ignored London [27]. He must have considered that there was no continuity from the gas to the liquid. It is now generally accepted that London was right, but, as we shall see, this issue is quite subtle.

Let us keep a few important steps only in the long history of this controversy. In 1947, Bogoliubov [28] showed that,

in a Bose gas with weak repulsive interactions, below the BEC transition, the low energy excitations are collective modes with a non-zero velocity. This is what allows such a weakly interacting Bose gas to be superfluid. Bogoliubov justified Landau's assumption that there are no single particle excitations at low energy, so that a superfluid current cannot dissipate its kinetic energy if its velocity is less than a critical value, here the velocity of this collective mode, which is identical to the ordinary sound velocity.

In my opinion, the next crucial step is the historical paper by O. Penrose and L. Onsager [29] who generalized BEC to an interacting system. The single particle states are no longer eigen states. They considered the one-body density matrix  $\rho_1(r)$  and its eigenvalues in the limit where  $r$  tends to infinity. They correspond to the occupation of the various eigenstates of the whole liquid. If the occupation of the ground state is macroscopic, meaning that the "condensate fraction"  $n_0$  is of order unity (when  $r$  tends to infinity), then BEC has taken place. Since the momentum distribution is the Fourier transform of this one-body density matrix, a finite  $n_0$  means a delta function at  $p = 0$  in the momentum distribution: there is a ground state with zero momentum which is occupied by a macroscopic fraction of the total number of particles. As explained by P. Sokol [30], the physical interpretation of  $\rho(r)$  is the overlap of a wavefunction of the system when a particle is removed at the origin and replaced a distance  $r$  away. The finite value of  $n_0$  indicates a long range coherence which is often called "ODLRO" for "Off diagonal long range order". The corresponding eigen function is the ground state wave function which is the order parameter for the transition. It is remarkable to see that, in the same article, Penrose and Onsager used Feynmann's wave function for the condensate, approximated liquid helium as a gas of hard spheres, and ended with  $n_0 \approx 8\%$  at low pressure, in agreement with more recent results. The ground state wave function has an amplitude and a phase  $\phi$ . The superfluid velocity is

$$v_s = \frac{\hbar}{m} \nabla \phi \quad (2)$$

More theoretical efforts have been done in order to calculate  $n_0$ . In his review [30], Sokol cites several numerical methods from "Path integral Monte Carlo" (PIMC) to "Green's function Monte Carlo" (GFMC) which are more or less consistent with the rough estimate first made by Penrose and Onsager. His GFMC predicts 9.2% at low temperature and pressure. As for measuring the condensate fraction in liquid helium it has been another challenge. Contrary to what happens with trapped Bose gases, where the condensate is directly observed and easily measured, there is no experimental evidence for the existence of a condensate in liquid helium. The quantitative analysis of "Deep Inelastic Neutron scattering" experiments is very delicate. Reliable experimental values for  $n_0$  were obtained in the last decade only. They depend not only on the assumption that a condensate exists, but also on the particular shape

of the distribution function for excited states with non-zero momentum. Given this “caveat”, the presently accepted experimental value for  $n_0$  in liquid helium at zero pressure and at low temperature is  $10 \pm 1.5 \%$ , in agreement with theory.

It is now interesting to consider the pressure (or density) dependence of the condensate fraction. As shown by Sokol [30] again, theory and experiment agree about this also:  $n_0$  decreases from about 10 % at  $P = 0$  bar where the density of liquid helium is  $\rho = 0.145 \text{ g.cm}^{-3}$  to about 5 % only at the melting pressure  $P_m = 25.3$  bar where  $\rho = 0.172 \text{ g.cm}^{-3}$  (Remember that liquid helium does not solidify at low temperature except if a pressure greater than 25.3 bar is applied). One understands the decrease of  $n_0$  as a result of the exchange between atoms becoming more difficult as the density increases in this range. A probably equivalent explanation is given by Bauer et al. [31] who write that “exchange is inhibited by the increase in effective mass of the particles”. This variation is consistent with the transition temperature  $T_\lambda$  being also a decreasing function of pressure. One now realizes that this behaviour is opposite to the prediction from Eq. 1. Does it mean that interactions always decrease the critical temperature for condensation? Certainly not, as shown by Gruter, Ceperley and Laloe [33]. Gruter et al. calculated  $T_{BEC}$  in a hard sphere fluid as a function of the parameter  $na^3$  which describes the interaction strength ( $n$  is the number density and  $a$  is either a scattering length in the dilute case or a hard core in the dense limit of liquid helium). They found that the variation of  $T_{BEC}$  is non-monotonic, a quite remarkable result. At low density, some interactions help the system to be homogeneous and to establish a long range coherence. On the contrary, large interactions make exchange difficult at large density. On Fig. 3, one sees that they were able to put the transition temperature of liquid helium on the same curve as their prediction for the critical temperature of BEC in dilute Bose gases. This curve would have surprised Landau !

I would like to make two further remarks on the pressure variation of  $T_\lambda$  in liquid helium. Suppose now that, by some clever mean, one could study liquid helium at a density lower than its equilibrium value. Would  $T_\lambda$  come closer to the ideal gas value  $T_{BEC}$ ? We have shown that this is possible by using acoustic pulses of high intensity and short duration [34]. In the absence of walls and impurities, it is possible to drive liquid helium to a metastable state at a negative pressure which approaches the “spinodal limit” at - 9.5 bar. Under such conditions, the density of liquid helium can be lowered to  $0.10 \text{ g.cm}^3$ , about 30 % less than in equilibrium at the saturated vapor pressure. Apenko [32] and Bauer [31] have calculated  $T_\lambda$  in this metastable region of the phase diagram (see Fig. 4). They both predicted that  $T_\lambda$  should reach a maximum value of about 2.2 K for a density of  $0.12 \text{ g.cm}^3$  corresponding to -8 bar.

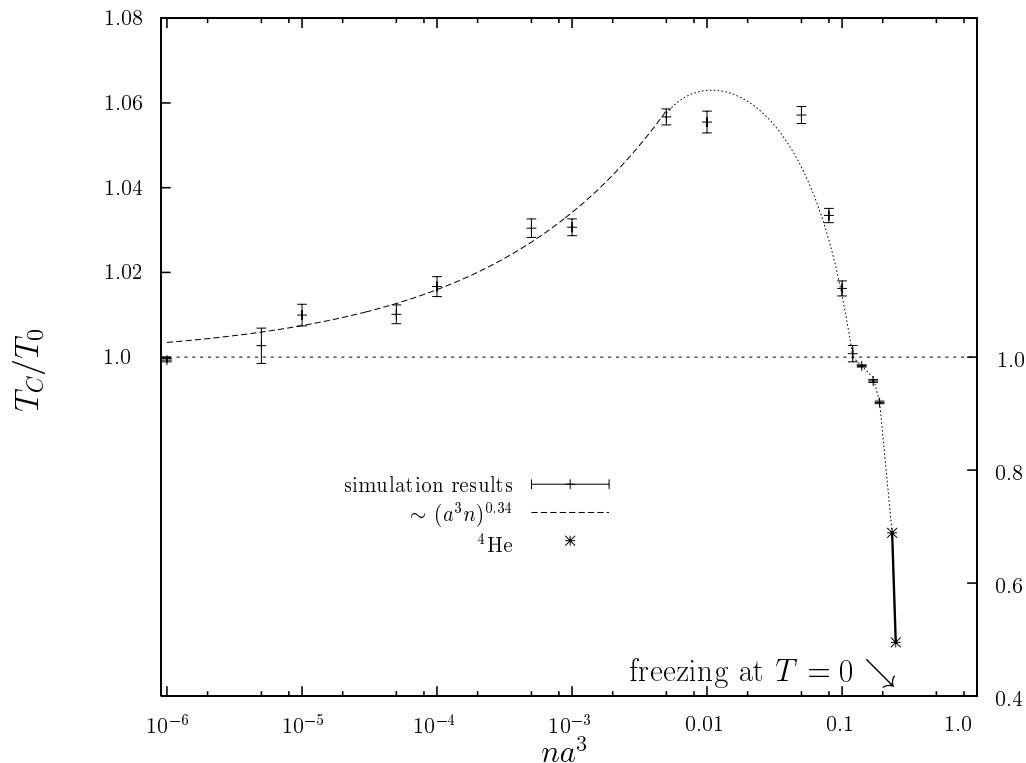


FIG. 3: The calculation by Gruter et al. of the BEC transition temperature in a hard sphere model as a function of the strength of the interactions. Note that the temperature scale is not the same for the low interaction part and for the large interaction part, so that an artificial cusp shows up where  $T_c$  crosses the value  $T_0$  of the ideal Bose gas. The star-like points on the right correspond to liquid helium. They lie on the same curve (dotted line) as for dilute gases.

In summary, in liquid helium at negative pressure,  $T_\lambda$  ceases to be a decreasing function of pressure and comes closer to the ideal gas behaviour. This is consistent with our observation of a cusp in the cavitation pressure at 2.2 K [34]. We naturally attributed this cusp to the crossing of the lambda transition at negative pressure, and we found it where it is predicted by recent theories, not on a linear extrapolation of the behavior at positive pressure. It is the sign that rotons are no longer dominating the thermodynamics of liquid helium near its spinodal limit.

We are also using the same acoustic technique to study overpressurized liquid helium. Our main motivation is to look for a possible limit of instability for the metastable liquid at high pressure with respect to the formation of the crystalline phase which is the stable one. In the course of this search, we have already reached about 120 bar without seeing nucleation of helium crystals. This shows that it is possible to study liquid helium at much higher densities than previously thought. In his review, Sokol [30] considers this region as “unaccessible” but he was not aware of our experiments. It is thus not absurd to consider what happens to superfluidity in liquid helium at high density. A crude extrapolation of Sokol’s calculation of  $n_0$  would predict that it becomes vanishingly small near a density of

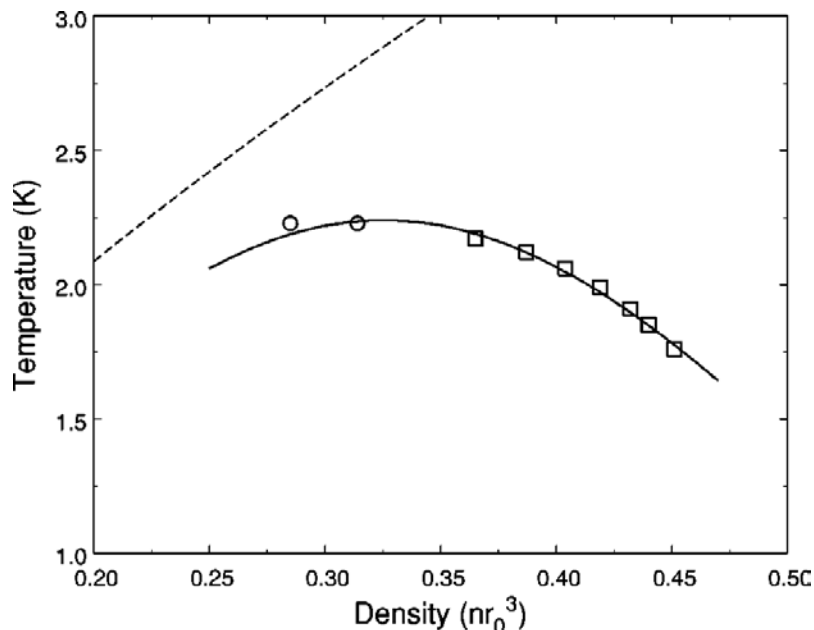


FIG. 4: The density variation of the lambda transition according to Apenko (solid line and symbols). The lambda temperature approaches the BEC temperature of the ideal Bose gas (dotted line) in the metastable region of liquid helium at low density. This behavior is consistent with the results obtained by Bauer et al.

about  $0.19 \text{ g.cm}^{-3}$ . Given the known equation of state [34], it would correspond to a pressure of order 55 bar, i.e. 30 bar above the equilibrium melting pressure. We have driven liquid helium up to a much higher density. It is hard to imagine that, if  $n_0$  is very small  $T_\lambda$  is not very small as well. How exactly does the lambda line extrapolate in this metastable region? Is it possible that a helium glass exists? It is certainly not easy to study such speculative properties but we might obtain some information on it from our experiments. It would also be interesting to calculate the properties of a quantum hard sphere model in the very high density limit where the system jams, not only in the very dilute case where the system approaches the ideal gas.

With these few comments and ideas, I hope that I have provided some information for a comparison between superfluid liquids and superfluid gases. My purpose was also to show that a few questions deserve further study.

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