Rotons, Superfluidity, and Helium Crystals

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Abstract. Fritz London understood that quantum mechanics could show up at the macroscopic level, and, in 1938, he proposed that superfluidity was a consequence of Bose-Einstein condensation. However, Lev Landau never believed in London's ideas; instead, he introduced quasiparticles to explain the thermodynamics of superfluid ^{4}He and a possible mechanism for its critical velocity. One of these quasiparticles, a crucial one, was his famous "roton" which he considered as an elementary vortex. AT the LT0 conference (Cambridge, 1946), London criticized Landau and his "theory based on the shaky grounds of imaginary rotons". Despite their rather strong disagreement, Landau was awarded the London prize in 1960, six years after London's death. Today, we know that London and Landau had both found part of the truth: BEC takes place in ^{4}He, and rotons exist.

In my early experiments on quantum evaporation, I found direct evidence for the existence of rotons and for evaporation processes in which they play the role of photons in the photoelectric effect. But rotons are now considered as particular phonons which are nearly soft, due to some local order in superfluid ^{4}He. Later we studied helium crystals which are model systems for the general study of crystal surfaces, but also exceptional systems with unique quantum properties. In our recent studies of nucleation, rotons show their importance again: by using acoustic techniques, we have extended the study of liquid ^{4}He up to very high pressures where the liquid state is metastable and we wish to demonstrate that the vanishing of the roton gap may destroy superfluidity and trigger an instability towards the crystalline state.

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This London prize lecture is an opportunity to review some aspects of superfluid helium, a macroscopic quantum system which surprises us since London's time. Rotons will be my guide because I wish to recall the controversy which opposed Fritz London to Lev Landau (Section 1). After this historical introduction, we shall see that Landau's famous quasiparticles are more or less present in most of my own work: quantum evaporation (Section 2), the surface of helium crystals (Section 3), and the stability limits of liquid helium (Section 4).

1. FRITZ LONDON AND LEV LANDAU

Fritz London arrived in Paris in 1937. He was trying to escape from the antisemitic Germany and Paul Langevin had offered him a position at the Institut Henri Poincaré. Langevin was a professor at the Collège de France and an influent member of the "Front populaire", the coalition of political parties from the French left. In January 1938, London had understood that the large molar volume of liquid helium was a consequence of the quantum kinetic energy of He atoms. He also realized that the superfluid transition temperature, the "lambda" point $T_\lambda = 2.17$ K, was close to the temperature at which an ideal gas with the same density would undergo a Bose-Einstein condensation (BEC). He eventually noticed that the singularity

in the specific heat at T_λ was similar to the one expected at a BEC transition, although not quite the same as could be expected since liquid helium is not an ideal gas. He thus proposed that superfluidity was a consequence of some kind of BEC[2].

One month later, Laszlo Tisza developed London's idea and introduced his "two fluid model"[3] to describe the non-classical properties of superfluid helium which had been discovered by J.F. Allen, in particular the fountain effect [1]. Laszlo Tisza had been in a Hungarian jail for 14 months after being accused of being a communist; after his liberation he went to Kharkov (1935-37) as a post-doc in Landau's group and finally arrived in Paris where he found help from the same group of left intellectuals around Langevin. He had a position at the Collège de France, 300 m from Fritz London. When London first heard about Tisza's two fluid model, he could not believe that, in a liquid which was pure and simple, there could be two independent velocity fields; this was indeed quite a revolutionary idea. But the joined works of London and Tisza explained most of the helium properties which were known in 1938.

At the same time in Moscow, Piotr Kapitza had invented the word "superfluidity" in analogy with superconductivity because he had the remarkable intuition that these two phenomena should have a common explanation [4]. As for Lev Landau, Stalin and Molotov had

put him in jail on the accusation of espionage. After a heroic fight, Kapitza succeeded in liberating Landau [5]. In 1941, Landau published the famous article in which he invented the concept of quasiparticle in quantum fluids [6]. There had to be two kinds of quasiparticles to describe superfluid ^{4}He, phonons and some others which he called "rotons" because he thought that they were elementary vortices. Together with phonons, rotons formed the normal component in a two fluid model which was similar to the one first introduced by Tisza. However Tisza had reasoned in the frame of gases and he thought that the normal fluid was made of the atoms left out of the condensate.

In his 1941 article, Landau was very critical about his former post-doc :

"the explanation advanced by Tisza not only has no foundations in his suggestions but is in direct contradiction with them"[6].

Furthermore, he never mentioned BEC and never referred to London. How could this be ? For a long time, I thought that Landau had an objection similar to the critic raised by London himself: the properties of an ideal gas could not apply to a liquid. We know how much work was necessary to extend BEC to interacting systems. More recently, a rather likely explanation came to me from a discussion with Lev Pitaevskii. The year 1941 was long before the BCS theory and the introduction of Cooper pairs. Since Kapitza and Landau thought that superfludity and superconductivity had the same origin, and since ^{4}He atoms were bosons while electrons were fermions, the quantum statistics could not be involved!

A few years later, when ^{3}He became available, it was crucial to see if liquid ^{3}He was superfluid at temperatures of order 1 K, also to verify if Landau's rotons had any reality. The first LT meeting took place at Cambridge (UK) in 1946, and the opening talk was given to Fritz London. He insisted on his explanation of superfluidity, also on the fact that Peshkov's early experiments on thermal waves (second sound) could not distinguish between Tisza's and Landau's predictions. He must have been also quite upset by Landau's attitude to comment on *"Landau's theory based on the shaky ground of rotons"* in the following way: *"The quantization of hydrodynamics [by Landau] is a very interesting attempt...however quite unconvincing as far as it is based on a representation of the states of the liquid by phonons and what he calls "rotons". There is unfortunately no indication that there exists anything like a "roton"; at least one searches in vain for a definition of this word...nor any reason given why one of these two fluids should have a zero entropy (inevitably taken by Landau from Tisza)"* [7]. Clearly, London and Landau had rather different approaches to superfluidity and had both easily recognized the weak points of their opponent's theory.

A few years later only, Osborne et al. [8] showed that ^{3}He was not superfluid down to 1 K and Peshkov found that the second sound velocity increased below 1 K, as predicted by Landau but not by Tisza [9]. Some more years later, the existence of rotons was demonstrated by neutron scattering experiments [10].

Both London and Landau probably died too early to admit that they both had part of the truth: BEC takes place in superfluid ^{4}He, also in ^{3}He and in superconductors thanks to the pairing of fermions, as for rotons, nobody doubts of their existence, it's only their physical nature which is still somewhat controversial, as we shall see below. One of my motivations in recalling this old time is the following message which I received from Laszlo Tisza himself, on the 17th of June 2005 :

Dear Sebastien,
I am delighted to read in Physics Today that you are to receive the Fritz London Prize. [...] This is wonderful! Please receive my warmest congratulations. Yesterday I was leafing through old correspondence and I found a letter in which I nominated Landau for the Prize. I am sure I was not alone. I was actually at LT-7 in Toronto when the Prize was announced. It is actually unconscionable of Landau not to have taken note of the remarkable Simon - London work on helium In Oxford 1934-5! I never heard a word of it while at UFTI. All he said that London was not a good physicist. I am looking forward to your book to straighten out matters. With warmest regards, Laszlo

The content of this message would need a lot more comments, but, for the present lecture, let me only wish Laszlo Tisza to enjoy many more years of scientific activity after turning 100 in 2007.

2. QUANTUM EVAPORATION

When I started playing with heat pulses in superfluid ^{4}He, I was in fact trying to detect the emission of vortices by a flow through an orifice. It was shown later by the group of Varoquaux and Avenel[11], followed by the group of Packard and Davis[12] that much more sensitive techniques were needed for this. But I was surprised to observe that heat pulses could propagate through the liquid-gas interface during the filling of our cell. After observing that, at high temperature, heat propagated as a second sound wave in the liquid and could emit ordinary sound in the gas when hitting the liquid-gas interface, I realized that the low temperature regime was much more interesting. The vapor pressure vanishes exponentially as T goes to zero, so that atoms propagate ballistically on macroscopic distances. Furthermore, the mean free path of phonons and rotons also becomes large, so that a heat pulse propagates as a burst of ballistic rotons and phonons. In such a regime, I heard from Horst Meyer who had worked on this phenomenon[13]

that P.W. Anderson[14] had predicted that it should be similar to the photoelectric effect. Quasiparticles incident on the liquid-vapor interface could evaporate atoms in a way similar to photons ejecting electrons from the surface of a metal. The conservation of energy would imply that the kinetic energy E_a of the evaporated atom would be equal to the difference between the quasiparticle energy and the binding energy E_b of atoms to the liquid. Since rotons have a minimum energy $\Delta = 8.65$ K and $E_b = 7.15$ K, atoms evaporated by rotons should have a minimum kinetic energy $\Delta - E_b = 1.5$ K. This corresponds to a minimum velocity $v = 79$ m/s.

By varying the liquid level in my cell and by measuring the flight time from a heater in the liquid to a bolometer in the gas, about 1 cm above, I could measure the velocity of atoms evaporated by rotons. I found the first preliminary evidence for the minimum velocity predicted by Anderson.[15] The photoelectric effect is the experimental evidence for the quantization of light. Similarly, my experiments showed that heat in a superfluid can be quantized as quasiparticles, especially Landau's famous rotons. Rotons are difficult to detect with a bolometer because their reflection probability on any solid surface is high. After being transformed into ballistic atoms, rotons were easy to detect and evidence for their minimum energy found.

Given these preliminary results, I asked Adrian Wyatt if we could continue together on this subject and use his experimental techniques which were much more sensitive than mine, in order to be more quantitative. Adrian called the phenomenon "quantum evaporation" and we got particularly interested in the case of anomalous rotons. This is because we expected the component of the momentum parallel to the surface to be conserved, as usual, not the velocity of course. Now, rotons have two branches on each side of the minimum in their dispersion curve: R^+ have their momentum parallel to their group velocity but R^- have it antiparallel. As a consequence, the evaporation by R^- rotons traveling to the right should evaporate atoms traveling to the left. For this, I started building a cell in Adrian's laboratory at Exeter, where heaters and detectors could rotate in a vertical plane, but my postdoc time ended long before this cell could be finished and work. In fact, Adrian Wyatt and his group worked on quantum evaporation for more than two decades and performed a beautiful analysis of the whole phenomenon: they not only observed evaporation by rotons but also by phonons, they could also separate evaporation from R^+ and from R^- rotons and obtain clear evidence for the anomalous evaporation we had imagined.[16, 17, 18] They also measured most evaporation probabilities by the various kinds of quasiparticles and compare with calculations by Dalfovo et al.[19]. This calculation included predictions on the reverse process, quantum condensation of atoms incident

on the free surface of liquid helium, which was first observed by D.O. Edwards et al. and depends on their momentum.[20] I am not sure that quantitative agreement is well established between theory and experiments on quantum evaporation and condensation and I have always been a little surprised to see that very few groups performed experiments on this. Among them is the one led by H.J. Maris and G. Seidel whose particle detection method involves quantum evaporation, and whose results on solar neutrinos might be of great importance.[21]

3. THE SURFACE OF HELIUM CRYSTALS

Together with Harry Alles and Alexandr Parshin, we have written a detailed review of this whole field which appeared this year. [22] I wish to focus here on some aspects of my own work only. I started studying the surface of helium crystals after noticing that, when crystallizing superfluid helium in a cell, the crystal position was sensitive to gravity: it occupied the bottom part as if it was water in a glass. David Edwards came to visit me in Paris and he suggested that we could try to measure the surface tension α of solid helium by measuring its capillary rise in a thin capacitor. We did this together [15] and obtained the first direct measurement of α, in fact an average value of the surface stiffness $\gamma = \alpha + \partial\alpha/\partial\phi^2$ (ϕ is the angular orientation of the surface) which governs the surface curvature in a generalized Laplace equation[22]. We also had two surprises. The capillary rise was negative, it was a depression because, apparently, the copper walls of our capacitor were preferentially wet by liquid helium. Moreover, we found an anomaly around 1 K: helium crystals looked much stiffer below this temperature than above. At the same time in Haifa, Jud Landau and Steve Lipson measured γ in their optical cryostat and found agreement with our results above 1 K but not below where facets showed up on the shape.[24] Also at the same time but in Moscow, Konstantin Keshishev, Alexandr Parshin, and Alexeï Babkin discovered that capillary waves could propagate at the surface of helium crystals below about 0.6 K as if it was a free liquid surface. They called them "melting freezing" or "crystallization" waves and obtained a value for γ at low T in agreement with our high temperature value but not with our low temperature value. I soon realized that, when facetted, a helium crystal could not pop through a hole as if it were a liquid, so that, below 1 K, our measurement was wrong. I also realized that, in order to study these crystals, it was extremely useful to see through the cryostat walls, that is to drill holes in the stainless steel and put windows.

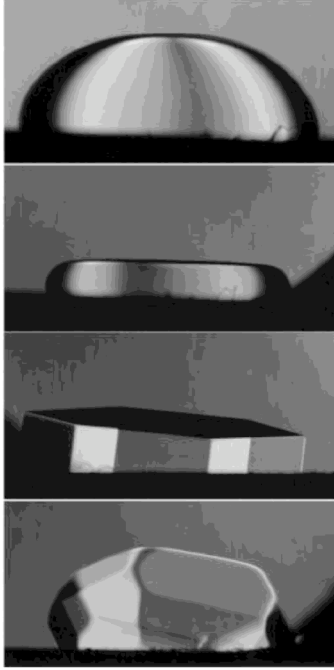In 1926, when Keesom discovered that superfluid

FIGURE 1. As temperature goes down, more facets appear at the surface of ^{4}He crystals.

helium solidified at 25 bar, he tried to observe the liquid-solid interface but he failed. Through the walls of his glass Dewar, "...there was nothing peculiar to be seen...". Some fifty years later, we had better techniques and the optical observation of this interface brought very interesting information (Fig.1). Among the numerous results obtained by many groups, I wish to mention here the roughening transitions and the crystallization waves.

3.1. The roughening transitions

Most of the static properties of helium crystals are common to all other crystals. A central one is the existence of successive roughening transitions where new facets appear on the equilibrium shape as T decreases; one says that the crystal surface changes from *rough* to *smooth* in order to express that its large scale fluctuations disappear (it is *not* a change at atomic scale). Thanks to a complete study of the first transition where so called "c" facets of the hexagonal structure appear, we have found precise agreement with the set of renormalization-group theories which predict that roughening transitions belong to the Kosterlitz-Thouless universality class. In particular, we have found that the step free energy vanishes exponentially as T approaches the roughening temperature T_R from below; we also found agreement with the universal relation between T_R and the surface stiffness

$$\gamma_R = \gamma(T_R)$$

$$k_B T_R = \frac{2}{\pi} \gamma_R d^2 \tag{1}$$

where d is the step height. We also found agreement for the critical variation of γ as a function of orientation, the critical variation of the growth rate as a function of temperature and growth driving force, and the critical behavior of the surface stiffness as a function of orientation and temperature. For a detailed review, please see ref. [22]. Here I only wish to insist on the universal relation (Eq.1) which is the best known property. Agreement was found between Nozières' theory[26] and all the results obtained by us and by the Moscow group [27]; for this we had to adjust three parameters, namely the roughening temperature T_R itself, the strength of the coupling of the interface to the underlying lattice, and a small scale cutoff where fluctuations start.

The c facet is the simplest one for such a comparison because ^{4}He crystals are easier to orient with a c facet horizontal than with any other facet horizontal, also because the c axis is a six-fold symmetry axis so that there is only one component of the surface stiffness tensor in this direction. The study of the other facets in ^{4}He is still incomplete so that the agreement with Nozières' theory is not as precise but all measurements are compatible with its predictions (see ref. [22]). As for ^{3}He crystals which are bcc, their (110) facets could only be seen below about 100 mK[28] although the roughening temperature was predicted to occur at 260 mK. This has been a puzzle for a long time. Thanks to the recent work of Todoshchenko et al. in Helsinki [29], one now understands that the coupling of the surface is extremely weak close to the roughening temperature, so that facets have a negligible size except if the temperature is much lower than 260 mK. A surprising result by the Helsinki group is that this coupling becomes strong at low temperature since quantum fluctuations of the surface are strongly damped when the viscosity of liquid ^{3}He is high.

Thanks to the study of helium crystals, the universal relation of roughening is now well established and we used it to explain the very large number of facets observed by P. Pieranski at the surface of some lyotropic liquid crystals[30]. These cubic crystals show up to 60 different types of facets at room temperature, which are arranged in sets of "devil staircases" around high symmetry orientations. Together with Nozières, we showed that these crystals are soft in the sense that their typical elastic energy is much smaller than their surface energy, so that their steps are in fact embedded as edge dislocations below the crystal surface. We could calculate their stiffness components and show that the large interaction between steps is compensated by the small step energy in the calculation of the roughening temperatures, so that the existence of a large number of facets at room temper-

ature is a simple consequence of the large value of the lattice spacing[31].

3.2. Crystallization waves

Contrary to most of their static properties which could be generalized to classical crystals, the dynamic properties of helium crystals are obviously particular to quantum systems. For example, around 100 mK, the growth rate of rough crystal surfaces is larger by 11 orders of magnitude in ^{4}He than in ^{3}He. No classical crystal would show such a difference between two isotopes. In ^{4}He, the growth rate increases as temperature goes down while, for classical crystals, everything depends on thermal activation above energy barriers, so that the growth rate always decreases when temperature goes down. The striking difference between ^{4}He and ^{3}He has two different origins. Firstly, excitations are different in superfluid ^{4}He and in a Fermi liquid such as ^{3}He. The crystallization of superfluid ^{4}He is mainly limited by collisions of the moving crystal surface with excitations; as a consequence, since thermal excitations disappear at low T, the growth rate diverges to infinity as T tends to zero.[32, 33] This is reminiscent of the mobility of electrons in metals which also increases as the density of thermal phonons vanishes. Since the excitations in liquid ^{3}He are Fermi quasiparticles which have a large momentum, the intrinsic mobility of the liquid-solid interface is much smaller than in ^{4}He (there is more momentum exchange during a collision with Fermi quasiparticles than during collisions with thermal phonons). As a consequence, even at 320 mK where the latent heat of crystallization is zero in ^{3}He, the shape of a ^{3}He crystal relaxes to equilibrium in a few seconds, which is about 6 orders of magnitude than in ^{4}He at the same temperature. But as soon as the latent heat is non-zero, since the thermal conductivity of liquid ^{3}He is poor, the resistance to growth involves a bulk thermal resistance which may be very large while it is negligible in ^{4}He. As a result, the growth dynamics of ^{4}He crystals looks strikingly fast while it looks as slow as for classical crystals in ^{3}He.

A famous consequence of the fast dynamics of ^{4}He crystals is the existence of crystallization waves. These waves are well defined on rough surfaces below about 0.6 K, when thermal rotons disappear and the growth rate is only limited by phonons. In these waves, the restoring forces are gravity and surface tension as for waves at a free liquid surface, and the kinetic energy comes from the mass transport which is necessary to change a liquid into a solid phase whose density is larger. As a consequence, one can use a measurement of their dispersion relation to obtain precise values of the surface tension (more precisely the surface stiffness). This proved particularly
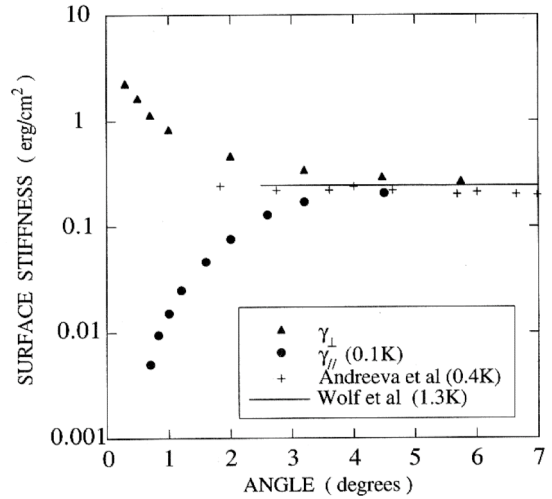

FIGURE 2. The extended phase diagram of helium. The liquid phase can be observed in a metastable state at negative pressure down to -9.5 bar and at high pressure up to 160 bar, possibly up to an instability line where the roton gap vanishes.[40].

interesting to study vicinal surfaces which are tilted by a small angle with respect to facets.

A vicinal surface is called "stepped" if it is made of well separated steps. For this the tilt angle has to be small enough. At small tilt angle, the vicinal surface properties are governed by step properties while at larger angle they are isotropic rough surfaces. With E. Rolley[34], we studied stepped surfaces to measure the properties of steps. For this, we built a cell which could rotate by $\pm$ 6° around two perpendicular axes. In this cell we first grew a crystal with a vertical c axis; it was wide enough for gravity to force the crystal surface to be horizontal. When rotating the cell, the surface kept horizontal but its crystalline orientation rotated. This allowed us to propagate waves either perpendicular to steps or parallel to them, also to vary the step density and of course the temperature. Fig. 3 shows some of the results of this delicate experiment. One sees that the surface stiffness becomes highly anisotropic as the tilt angle ϕ tends to zero. We verified that the component $\gamma_{//}$ of the stiffness tensor diverged as $1/\phi$ while the component $\gamma_\perp$ was proportional to ϕ. A stepped surface is somehow like a corrugated board, very soft to bending in one direction but very stiff in the other direction. From a fit with theory, we obtained the step energy and the interaction between steps which had two parts, an entropic repulsion and an elastic one. We could also estimate the step width from the crossover angle (about 2.5°) from rough to stepped behavior and we found very good agreement with what we had already learned when studying the roughening transition. We also understood
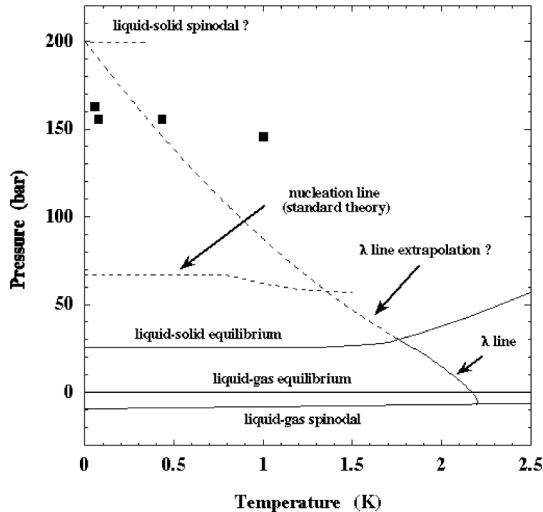
FIGURE 3. The anisotropy of the surface stiffness of ^{4}He crystals, as measured by Rolley et al.[34]

the details of step-phonon collisions from the damping of crystallization waves on stepped surfaces, we obtained evidence for the adsorption of ^{3}He impurities on these steps, etc.

At the end of this long study, I could compare our results on He steps with those obtained by Ellen Williams and her group on Si crystals. At high enough temperature on vicinal surfaces of Si crystals, the steps fluctuate, consequently the width of terraces between them. E. Williams had measured the distribution of terrace widths and found it narrower in the presence of the elastic repulsion.[35] Our description of step-step interactions was perfectly consistent with hers and this convinced me of the great interest of helium as a model system: with helium crystals one has access to some physical quantities which are not easy to measure in more classical systems but the information obtained in helium can be generalized to all others.

4. THE STABILITY LIMITS OF LIQUID HELIUM

In the recent years, we have extended the phase diagram of liquid helium to negative pressure where it is metastable with respect to the gas and to high pressure where it is metastable with respect to the solid. Thanks to acoustical techniques which I learned from Humphrey Maris, we are now able to study superfluid ^{4}He from -9.5 bar up to 160 bar, possibly more, a pressure range much larger than the stability one (0 to 25 bar). This is because the liquid-gas and the liquid-solid transitions are first order, so that the nucleation of the stable phase oc-

curs a certain distance away from the equilibrium line. If there are no impurities and if one eliminates the influence of walls, no "heterogeneous nucleation" takes place, the nucleation has to be "homogeneous". Our experiments use piezoelectric transducers with a hemispherical shape. They are excited at resonance in a thickness mode, so that bursts of ultrasound (typically 1 MHz) can be focused at the center of the transducer, far from any wall.

One difficulty is the calibration of the pressure amplitude at the acoustic focus. By studying the variation of the nucleation threshold as a function of the static pressure in the cell, we could measure this amplitude within about 10 percent. We found that, in the low temperature limit, bubbles nucleate, i.e. cavitation occurs near the spinodal limit which has now been calculated by several methods as -9.5 ± 0.2 bar near $T = 0$ in ^{4}He.[36] We also found a crossover from a quantum nucleation regime below about 0.2 K to a thermally activated one at higher temperature. In ^{3}He, we showed that cavitation occurs near -3 bar and that quantum cavitation is not possible in a short time there because of the existence of zero sound whose velocity does not vanish at the spinodal limit.[37] All these results and some others are discussed in a review article which I wrote on "Nucleation in quantum liquids" [38].

More recently, we developed our research in two different directions. One is the extension to liquid water of acoustic cavitation. This is another example of the model character of helium: we are extending what we learned in helium, a simple liquid whose spinodal line is well established, to THE complex liquid where the spinodal line is a matter of controversy and where homogeneous cavitation studies can distinguish between two competing models of its structure. The other direction is the homogeneous crystallization threshold of superfluid helium. For this we use the same type of acoustic technique but we study nucleation in the positive swings of the waves instead of the negative ones. This will be my last subject for this lecture, and, as we shall see, it will take us back to rotons.

In a first series of experiments, we focused the acoustic waves on a clean glass plate.[39] From the reflectivity at the glass-helium interface, we could measure the local density, or the local pressure by using the known equation of state of liquid helium. We found crystal nucleation 4.3 bar above the liquid-solid equilibrium pressure $P_m = 25.3$ bar (see Fig. 4). This was 2 to 3 orders of magnitude larger than in previous experiments where favorable defects or impurities must have been present on cell walls. This experiment also showed that crystals could grow at 100 m/s, reach a 10 μm size in 100 ns and be easily observed. Eventually, we studied the statistics of nucleation and showed that the nucleation was heterogeneous, taking place on one particular defect of the glass plate.
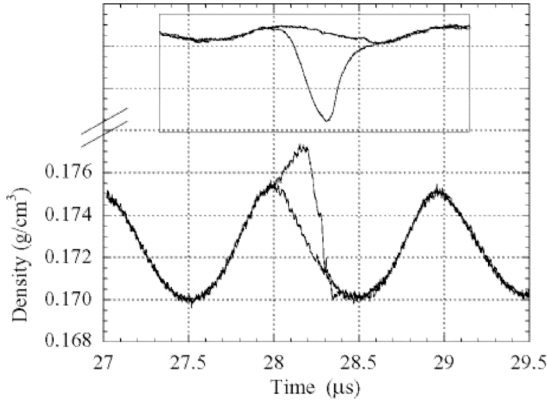
FIGURE 4. Acoustic nucleation of ^{4}He crystals occurs 4.3 bar above $P_m = 25.3$ bar, the liquid-solid equilibrium pressure. One records the density at the acoustic focus from the light reflected at the glass-helium interface (lower traces); the nucleation is also easily detected from the transmitted light (upper traces). At the threshold, one observes either one or the other type of signal with a probability 0.5. [39].

In a second series of experiment, we removed the glass plate.[40] Apparently, we observed cavitation in the negative swings but no crystallization in the positive swings, even at 25 bar near P_m. This was a surprising result which showed that the standard nucleation theory could not be used for precise predictions far from equilibrium. Indeed, it uses the "thin wall approximation" to calculate the nucleation barrier with the value of the macroscopic surface tension of crystals in equilibrium with liquid helium. This elementary calculation predicts homogeneous nucleation at 65 bar while we could overpressurize liquid helium up to 160 bar without nucleation. According to Maris and Caupin [41], the liquid-solid interfacial energy increases with density so that the standard theory underestimates the nucleation barrier.

How far can one pressurize liquid helium before it crystallizes? This is in fact a rather open question. In 1971, Schneider and Enz proposed the existence of a stability limit where the roton gap Δ vanishes.[42] Δ is known to decrease with pressure from 0 to 25 bar. If Δ vanishes, rotons become a soft mode which triggers an instability: a density wave should spontaneously grow and break the translation symmetry of the liquid. Since the wavevector of rotons is the inverse of the interatomic distance, the periodic phase is likely to evolve into the stable crystalline phase. H.J. Maris estimated that $\Delta = 0$ around 200 bar from an extrapolation of Dalfovo's density functional.[43] More recently, it was found from Monte Carlo simulations that this stability limit probably occurs at even higher pressure.[44]

In my opinion, this instability illustrates the nature of rotons. Landau chose this name because he thought that they could be elementary vortices. Later, Feynman modified Landau's view and tried to consider rotons as elementary vortex rings, but this new image had a difficulty.[45] Indeed, the group velocity of rotons with energy Δ is zero, and this is hardly compatible with the image of a vortex ring. Nozières recently noticed [46] that the existence of a roton minimum in the dispersion relation $\omega(q)$ of helium excitations is just a sign of local order in liquid helium. Feynman himself explains that $\omega(q)$ is proportional to the inverse of the structure factor $S(q)$, so that the roton minimum is a consequence of the existence of a large maximum in $S(q)$ (a "Bragg peak" as Nozières says). There is some local order in liquid helium, consequently a large probability to find atoms at an atomic distance from another atom, and the dispersion of phonons with a wavelength equal to the interatomic distance resembles that of a periodic crystal. As pressure increases, the local order increases and the roton minimum tends to zero. In this representation, rotons are density fluctuations signaling the proximity of a crystalline phase. When the roton minimum vanishes, the liquid becomes unstable. This is the new view of rotons which we hope to verify experimentally.

With this in mind, we have started a third series of measurements, now with two hemispherical transducers forming a spherical one. The amplitude of positive peaks is higher but the calibration is more difficult because the acoustic focusing is more non-linear than in the hemispherical geometry.[47] According to our preliminary results [48], we have found homogeneous nucleation of ^{4}He crystals in the bulk of liquid ^{4}He, a long standing challenge.

In order to measure the nucleation pressure, we plan Brillouin scattering measurements inside the acoustic wave. We should obtain the local instantaneous sound velocity and relate it to the local pressure from the most recent equation of state [44]. In fact, we expect Brillouin scattering to tell us about another important issue, namely the vanishing of superfluidity as a function of pressure. Superfluidity is a long range quantum order which requires exchange between atoms. The higher the pressure, consequently the density, the more difficult is this exchange. This is why the superfluid transition temperature decreases as a function of density, contrary to the BEC transition in a weakly interacting gas. We wonder what happens to superfluidity in highly pressurized liquid ^{4}He, how the lambda line extrapolates at densities of order 0.23 g/cm^3, 30 percent more than at 25 bar. At first sight, we expected the lambda line to join the liquid-solid instability line at $T=0$, as drawn on Fig. 2. But Nozières' argued that superfluidity could disappear before Δ becomes zero.[46] Since Brillouin scattering allows to detect superfluidity from the existence of second sound, we plan to get some information on this issue. If we could perform Raman scattering as well, we could perhaps also measure the vanishing of the roton

gap. Such experiments look difficult but worth trying in the coming years.

ACKNOWLEDGMENTS

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