Metastable Phases of Liquid and Solid 4 He

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Metastable Phases of Liquid and Solid ^{4}He

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Abstract

Experiments and theories describing the metastable phases of liquid and solid ^{4}He are presented and discussed. For the case of metastable liquid ^{4}He with respect to its gaseous phase, it is shown that different measurements of its destabilization threshold (cavitation threshold) and their comparisons to available theories reveal that the nucleation mechanism is not totally understood. Then experiments measuring typical lifetime of cavitation bubbles in He I and He II are shortly considered showing the important role of heat transport mechanism during their collapse. Finally for liquid ^{4}He, its metastability with respect to its solid phase and the possibility of the liquid destabilization due to the vanishing of the roton gap is presented. The last part of the review is devoted to metastable solid ^{4}He with respect to its liquid phase. The first experimental production of such a state is described and its destabilization limit possibly invoking the creation and proliferation of crystalline defects is discussed.

1 Introduction

^{4}He at low temperature is a model material for condensed matter physics mainly for two reasons. First, atom interactions are weak and quantum effects play an important role in understanding the macroscopic properties of the sample. Second, condensed ^{4}He can be obtained with a remarkable purity. When preparing a sample of liquid or solid ^{4}He in an experimental cell, atomic or molecular impurities other than helium are frozen in the filling line of the cell or on the walls of the cell itself. It is then expected that intrinsic helium phase properties can be studied without being modified by the presence of impurities.

Achieving a metastable phase of (for instance) liquid ^{4}He consists in bringing a sample of liquid ^{4}He at a temperature and a pressure (or density) where the thermal equilibrium phase is not the liquid phase but rather the solid one or the gaseous one. The possibility of having metastable liquids or solids is due to the fact that the solid/liquid and liquid/gas transitions are first order phase transitions. The density of the two phases involved in the transition is discontinuous at the transition. The interface between the lower and higher density phases has a given energy per unit area (the surface tension in the case of a liquid/gas mixture). This interface energy dominates the bulk free energy gain for a small nucleus of one of the phase in the other. This produces an energy barrier for the growth of the nucleus and makes metastable states possible.

Usually, the nucleation of the stable phase within the metastable one occurs on "defects "such as impurities or walls of the container which locally lower the energy barrier. In this case the nucleation is said to be heterogeneous. On the contrary, when the nucleation is not triggered by defects, it is said to be homogeneous. Extremely pure liquid and solid ^{4}He appear then to be the ideal candidates for testing homogeneous nucleation theory.

In this short review, I shall discuss experiments and theories describing the metastable phases of bulk liquid and solid ^{4}He. Metastable phases of condensed ^{4}He confined in porous media have been investigated with specific detection methods[1, 2, 3]. One may question however the homogeneity of the physical conditions of the studied phases and the role played by confinement on the metastable phase properties. In any case, we discuss here only bulk metastable phases. A very important tool in order to experimentally produce metastable samples of bulk ^{4}He has been developed 30 years ago by *Nissen et al*[4] for the case of liquid ^{4}He. A piezo electric transducer is immersed in a cell filled with liquid ^{4}He (phase 1 in Figure 1). It creates a focused pressure/density wave within the liquid and bring it in a metastable state (point B of Figure 1). The essential advantage of this acoustic method is that it can produce large pressure/density swings far from any interface (walls of the container and of the transducer) and in a small volume (the acoustic focus) of the bulk of the sample. These two points strongly reduce the chance of generating heterogeneous nucleation events and hence make possible a deep exploration of the metastable state up to a point where homogeneous nucleation is expected to be observed. The main difficulty of the method resides in the measurement or estimation of the pressure (or density) at focus.

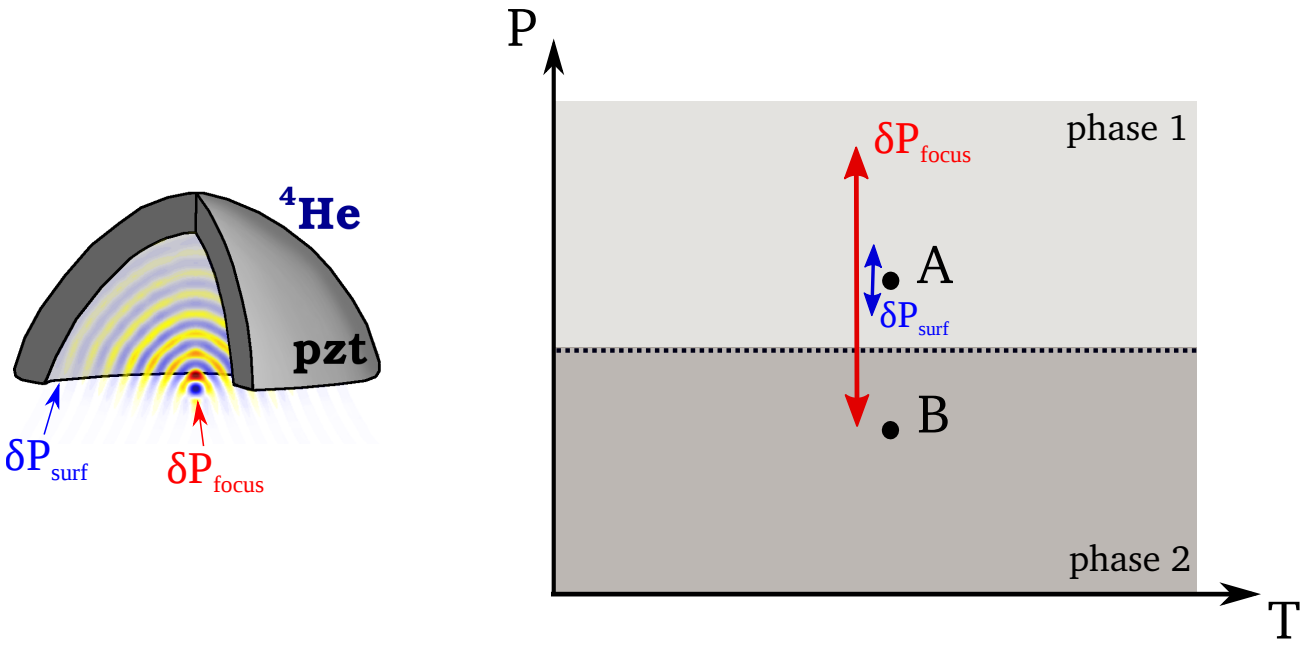

Figure 1: Principle of the acoustic technique used to obtain metastable states of condensed ^{4}He. A sample of liquid or solid ^{4}He is initially set at thermal equilibrium at point A of the phase diagram. An appropriate piezo electric transducer (pzt) creates a focused pressure/density wave within phase 1. The pressure variations at the surface of the transducer are small enough not to cross the coexistence line and heterogeneously nucleate phase 2 at the transducer surface. As the wave propagates, the pressure/density variations increase and ^{4}He in phase 1 can explore a part of the phase diagram where the thermal equilibrium phase is phase 2. Condensed ^{4}He in phase 1 at point B is in a metastable state.

As I shall recall in the first part of this review, this technique has been used by different groups during the 90s and early 2000s to produce liquid ^{4}He under tension. These experiments have triggered different theoretical works aiming at determining the properties of metastable liquid ^{4}He. Among these were the computation of, the equation of state of of liquid ^{4}He at negative pressures, the homogeneous cavitation threshold and the so called spinodal limit (which will be defined shortly after). In 2002, the agreement between the theory and the experimental results measuring the cavitation pressure of liquid ^{4}He suggested that the cavitation mechanism in metastable liquid ^{4}He was well understood. But we will see that more recent experiments on metastable liquid ^{4}He have questioned this conclusion and also questioned the validity of previous estimates of the cavitation pressures or the validity of the theoretical equation of state. I shall then discuss a new interesting property of the dynamics of cavitation bubbles in He I and He II. Finally regarding metastable liquid ^{4}He, I will consider experiments aiming at producing metastable liquid ^{4}He with respect to the solid phase (overpressurised liquid ^{4}He) where an interesting destabilization scenario of the liquid could take place.

The second part of this paper will be devoted to the metastable phases of solid ^{4}He. I will present how the acoustic technique has been adapted to the case of hcp solid ^{4}He where sound velocity is anisotropic in order to produce metastable low density states of the solid. The physical interest of metastable solid ^{4}He is again its extreme purity and the *a priori* possibility of testing homogeneous nucleation theory in a quantum solid. As we shall see, experimental results regarding the stability limit of the solid suggest that crystalline defects could play an important role in the destabilization of the solid.

2 Metastable Liquid ^{4}He

2.1 Liquid ^{4}He under tension

The situation in 2002 . The situation back in 2002 regarding cavitation in liquid ^{4}He is pretty well summarized in Figure 2 coming from the review "Nucleation in Quantum Liquids" written by S. Balibar and published in JLTP in December 2002[5]. On this Figure, the results of four different groups who had succeeded in producing high degree

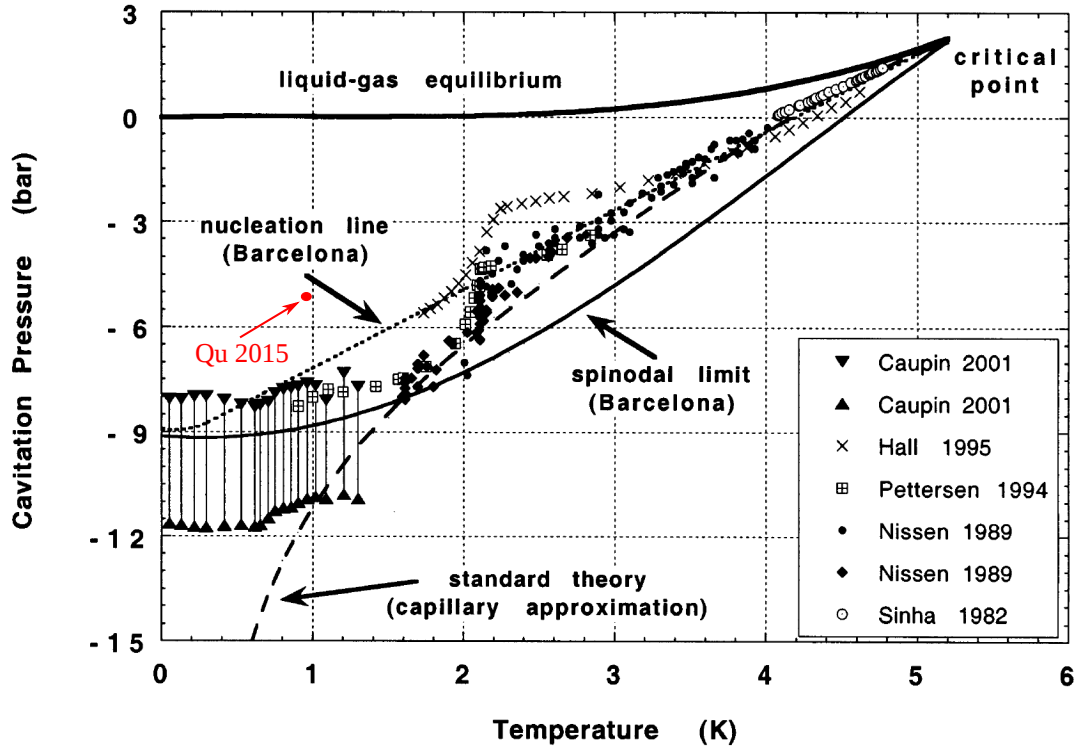

Figure 2: Summary of cavitation results in liquid ^{4}He back in 2002 taken from the review "Nucleation in Quantum Liquids" written by S. Balibar and published in JLTP in December 2002[5]. The cavitation density measurement converted into pressure using a theoretical equation of state of metastable liquid ^{4}He (see text) performed by Qu *et al.*[13] in 2015 has been added to the original figure.

of metastable liquid helium-4 and in generating cavitation in it are shown[1]. Apart from Sinha *et al.*'s experiment who superheated the liquid to obtain the metastable state [6], all other experimental results shown in the Figure were obtained using the acoustic technique. Nissen *et al.*[4] (who introduced this technique) used a relative indirect method to estimate the pressure at which cavitation occurs in the liquid. It is based on the computation of the pressure at acoustic focus knowing the geometry and the acoustic power of the emitter. Similar methods to estimate the cavitation pressure are employed by Hall *et al.*[9] and Pettersen *et al.*[10]. However, in these experiments, the sound propagation is highly non-linear and it is very difficult to correctly estimate the pressure at acoustic focus. To overcome this difficulty, Caupin *et al.*[11] used another method to estimate the cavitation pressure. They measured the voltage V_C applied to the transducer in order to observe cavitation as a function of the static pressure of the cell P_{stat}. The authors claim that, due to the non linearities in the sound propagation, the linear extrapolation of the curve[2] $(P_{stat}, \rho_{stat} V_C)$ to $V_C = 0$ gives an upper bound of the cavitation pressure[12]. They are also able to set a lower bound to the cavitation pressure by neglecting the non-linearities.

Three important theoretical results concerning metastable liquid ^{4}He are also shown in Figure 2. The first one labeled "standard theory"gives the prediction of the homogeneous nucleation of vapor bubbles (cavitation) pressure in liquid ^{4}He in the framework of classical statistical physics[14]. This prediction gives a homogeneous cavitation pressure which diverges as $-1/\sqrt{T}$ as $T \to 0$ and is not in agreement with experiments. Indeed, the standard theory of nucleation does not take into account the existence of the **spinodal limit** of the metastable liquid, which is the point where the metastable liquid is mechanically unstable even in the limit $T \to 0$. In other words, a real liquid can not support infinite tension without breaking. This spinodal limit in the case of liquid ^{4}He has been computed by different groups[15, 16, 17, 18] using different methods giving results which all are consistent with each other. The spinodal pressure $P_{sp}(T)$ at temperature T is defined by the equation $\left(\frac{\partial P}{\partial \rho}\right)_{T,P_{sp}} = 0$. The value computed by

[1] Two other groups have obtained similar results than Sinha *et al.*[6] in the same temperature range, these are Semenova *et al.*[7] and Nishigaki *et al.*[8]

[2] ρ_{stat} is the static density of liquid ^{4}He at static pressure P_{stat}

Jezek *et al.* using a density functional (DFT) approach[15] and labeled "spinodal limit (Barcelona)"is displayed in the Figure. In this paper, Jezek *et al.* were also able to compute the cavitation pressure of liquid ^{4}He in the DFT framework which is certainly an improvement to the standard theory of nucleation as the cavitation pressure tends to the spinodal limit as $T \to 0$.

In summary, back in 2002, the situation regarding metastable liquid ^{4}He and cavitation was the following. Experiments have indeed provided evidences for high degree of metastability of liquid ^{4}He. Measuring accurately the pressure at which the liquid cavitates is a difficult task for experimentalists. In the temperature range of He I, there is a fair agreement between the calculated homogeneous nucleation line and the experimental estimation of the cavitation pressure. In He II however, the experimental estimation of the cavitation pressure notably deviates from the theoretical nucleation line and seems to be compatible with the theoretical spinodal limit of the liquid. This later conclusion however must not be regarded as definitive, since recent experiments measuring the cavitation threshold density at 1 K have questioned it.

Cavitation density of liquid ^{4}He at 1 K. In our group, we have developed an apparatus to measure the density at which liquid helium ^{4}He cavitates. The metastable state is produced using the acoustic technique (1 MHz resonance) described earlier. Helium density modulations are monitored via the refractive index modulations imposed by the acoustic wave. Refractive index variation maps are measured using an optical Jamin interferometer (Fig. 3). From them, local density maps are simply obtained by assuming the small polarizability limit ($n - 1 \ll 1$) of the Clausius-Mossotti relation $\dfrac{\delta n}{n_0 - 1} = \dfrac{\delta \rho}{\rho_0}$. Note that the method is valid under the only assumption that the sound field has a symmetry axis. The density measurement does not depend on any assumption concerning the sound intensity and whether the material is or is not in the linear elasticity regime. This method of determining fluid density maps within a sound field has been quantitatively tested in water by comparison with an independent density measurement using an optical fiber hydrophone[19]. Deviations of less than 5% were found between the two methods. Hence this measurement of a liquid density in its metastable state provides quantitative data.

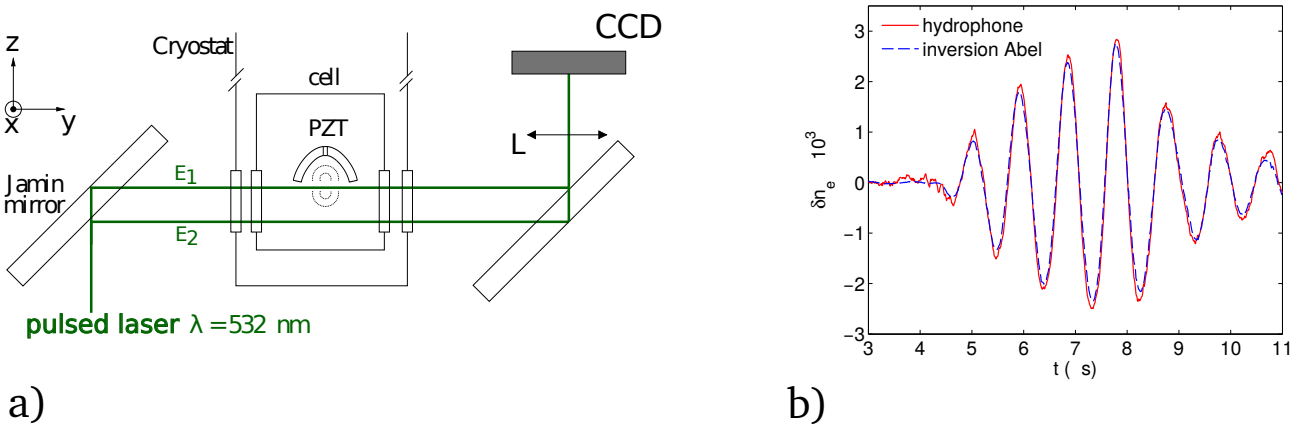

Figure 3: Apparatus used to measure the density at which liquid helium ^{4}He homogeneously cavitates. A Jamin interferometer is used to measure the optical phase shift induced by the acoustic wave on the optical beam E_1. The optical beam E_2 is not affected by the acoustic wave. The duration of the pulsed laser is 8 ns which is significantly smaller than the $\sim$ 1 μs period of the sound wave. Consequently an "instantaneous"image of the acoustic field is taken by the CCD camera. b) Refractive index variation temporal evolution $\delta n_{focus}(t)$ at acoustic focus induced by an acoustic wave focusing in a water. Red Curve : hydrophone measurement. Blue dash Curve : Our interferometric method based on Abel inverse of phase map profiles. The agreement between the two measurements strongly support that our method leads to reliable quantitative data.

Applying this method to determine the density of metastable liquid ^{4}He at the homogeneous cavitation threshold ρ_{cav} at $T \sim 1$ K, we found that $\rho_{cav} = 0.1338 \pm 0.0002\,\mathrm{g.cm^{-3}}$ [20, 13]. Within the error bars of our experiment, ρ_{cav} is found to be independent of the static pressure of the cell[13]. This value of ρ_{cav} turns out to be quite interesting when compared with previous works. As I already mentioned at the beginning of this chapter, other groups measured the cavitation pressure of liquid ^{4}He instead of the cavitation density. The equation of state (EOS) of liquid helium in its metastable state (density and pressure below the boiling curve values) is needed to convert the ρ_{cav} value to a corresponding P_{cav} value. The theoretical EOS of metastable liquid ^{4}He has been established by different approaches giving similar results[17, 21, 22, 23]. As H. Maris pointed out, in the stable phase at $T = 0.1$ K, the sound velocity

pressure dependence could be fitted very well by the law $c^3 = b(P - P_c)$ with c the sound velocity, P the pressure, P_c the spinodal pressure and b a constant [21]. He proposed that this relationship holds in the metastable state (negative pressure). Bauer *et al.* have performed Path-integral Monte Carlo simulations of liquid helium in the metastable state at finite temperature and found the same dependence of sound velocity on pressure[17]. Dalfovo *et al.* have calculated the EOS of metastable liquid helium at $T = 0$ K using density-functional approach[22], and Boronat *et al.* using a quadratic diffusion Monte Carlo method to achieve a similar EOS[23]. The EOSs at 0 K agree within a few percent. Moreover, using the density-functional theory of Dalfovo *et al.*, Maris and Edwards have shown that in the temperature range $0 < T < 1$ K, the EOS is nearly independent of temperature[24].

The value of our measurement of the cavitation density at $T \sim 1$ K can then be converted to a cavitation pressure. We surprisingly found $P_{cav}(1\text{K}) = -5.1 \pm 0.1$ bar. This value is far from the theoretical value of spinodal limit $P_{sp}(1\text{K}) \sim -9.0$ bar[15, 17] and is clearly not compatible with the one estimated by the extrapolation method of Caupin *et al.* $P_{cav}(1K) < -7.5$ bar as shown in Figure 2. One way to solve the disagreement between these two values would be to show that the theoretical EOS is not accurate and that a better one would reconcile the two measurements. For that, it appears necessary to measure experimentally the EOS of metastable liquid helium. It could be obtained by measuring simultaneously the density and the compressibility (that is the sound velocity) of the liquid in the metastable state. Such an experiment, using a stimulated Brillouin spectroscopy technique[25, 26] is currently being set up in our laboratory. If the theoretical EOS of liquid ^{4}He is confirmed experimentally, the difference between the two measurements must originate from somewhere else. For instance, the dependence of the cavitation voltage V_C on the static pressure P_{stat} could be more complicated than assumed by Caupin *et al.*.

Finally, even if we assume that the theoretical EOS is valid so that -5.1 bar is an acceptable value of the cavitation pressure at 1 K, it is not in agreement with the homogeneous nucleation theory of Jezek *et al.* which gives -6.9 bar (see Fig. 2). The possibility that nucleation in these kind of experiments is somehow heterogeneous should also be considered.

2.2 Heterogeneous cavitation of liquid ^{4}He

The possibility of heterogeneous nucleation of liquid ^{4}He is a rather tricky problem. As mentioned in the introduction, liquid ^{4}He is an extremely pure liquid as atomic or molecular impurities other than helium are frozen in the filling line of the experimental cell or on the walls of the cell itself. Isotopic ^{3}He impurities are present at the standard level of 0.3 ppm. They are unlikely to play a significant role in cavitation because of their atomic size, much smaller than the size of the critical nucleus for cavitation[27]. However, in the case of liquid He II, it was suggested that quantized vortices could play the role of impurities and lower the cavitation threshold. Using a DFT approach at zero temperature, Dalfovo has shown that at a critical negative pressure of -8 bars, a vortex line becomes unstable against free expansion of its core[28]. This corresponds indeed to a reduction of the cavitation threshold as the spinodal pressure at T=0 in the absence of vortices is about -9.6 bars[15, 17, 21, 22] (see Fig. 2). Maris has extended the Dalfovo calculation to finite temperature[21]. For a vortex density ranging from 10^4 to $10^{12}\,\text{cm}^{-2}$, he founds that the cavitation pressure assisted by vortices lies between -5.8 and -5.1 bar at T=1 K. This is indeed in the range of the cavitation density measurement[13] converted to pressure exposed in the previous section so that, if the theoretical EOS of liquid ^{4}He is confirmed experimentally, this measurement and the Maris calculation could be interpreted as an evidence for vortex assisted cavitation in liquid ^{4}He. However, as it can be seen in Figure 2, different acoustic experiments have estimated the cavitation pressure of liquid ^{4}He below and above the λ-point[4, 9, 10]. All display the common feature that the estimated cavitation pressure is less negative when going from the superfluid phase to the normal phase. This doesn't seem compatible with the vortex assisted cavitation scenario mentioned above. An interesting but rather hypothetic possibility to reconcile this scenario with these measurements is to invoke the Kibble-Zurek mechanism[29]. According to this mechanism, a "fast"quenched of a sample of liquid ^{4}He through the second order normal/superfluid transition creates within the liquid a tangle of quantized vortices[30]. If this mechanism is actually taking place in liquid ^{4}He, a possibility is that acoustic cavitation experiments performed at temperature above the λ-point could generate an important number of vortices by crossing the λ line (potentially in the metastable domain) which then lower the vortex assisted cavitation pressure threshold with respect to experiments performed at temperature below the λ-point where no crossing of the λ line did occur. I should insist on the fact that this idea is highly speculative as no quantitative theory is supporting it for now. My point is mostly to stress that looking carefully at cavitation thresholds at the λ transition both theoretically and experimentally could be an interesting prospect in the experimental search for a signature of the Kibble-Zurek mechanism in liquid ^{4}He. Until now indeed, the search for an experimental signature the Kibble-Zurek mechanism in liquid ^{4}He has been restricted to look for attenuation of second sound pulses with no evidence so far for the occurrence of the Kibble-Zurek mechanism in liquid ^{4}He [31, 32, 33].

If there is a relatively simple physical picture in which one can understand how vorticity can locally reduce the pressure and hence lower the cavitation threshold, there is a priori not such a simple picture for how surfaces (walls)

may facilitate nucleation in the case of liquid ^{4}He. The reason is that liquid ^{4}He perfectly wets surfaces made of any material with the noticeable exception of cesium[34]. Liquid ^{4}He is more attracted by any surface than by itself and walls are not expected to facilitate cavitation since creating a critical germ on a surface is not energetically favorable. However there are experimental evidences that heterogeneous nucleation making the cavitation pressure less negative does indeed occur in liquid ^{4}He. Regarding this issue, an important experiment has been conducted by Chavanne *et al.*[35]. The authors used the hemispherical transducer acoustic technique but inserted at acoustic focus a clean glass plate. Their observation was that cavitation pressure (T=200 mK) was lying in the range 0 to -3 bar, significantly less negative than in the case of homogeneous nucleation (no glass plate). The authors interpreted that as an evidence for heterogeneous nucleation on the glass plate. The group of L. Skrebk in Prague (Czech Republic) uses a vibrating quartz tuning fork to produce metastable states of liquid ^{4}He. Such a fork can be seen in Figure 4b). The authors observed cavitation both visually and electrically as a breakdown of the resonance response of the fork[36, 37]. From the location of the cavitation bubble in the vicinity of the fork prong and the estimation of the cavitation pressure (using the Bernouilli law) of -0.2 bar at He II temperatures, the authors conclude that they also observe heterogeneous cavitation in liquid ^{4}He.

Heterogeneous nucleation of liquid ^{4}He on solid surfaces has indeed been observed experimentally. For now, there is no real understanding on the physics involved and hence no theoretical model able to account for experimental results has been proposed. An interesting thing to look at for theoreticians would be to check if the roughness of a surface on the nm scale in "lobster pot" arrangements would efficiently lower the cavitation pressure as suggested in [36].

2.3 Lifetime of cavitation bubbles in He I and He II

Regarding low density metastable states of liquid ^{4}He, an interesting property which has recently come to light is the typical lifetime of cavitation bubbles. As we have seen, the acoustic arrangement schemed in Figure 1 is suitable to trigger a cavitation bubble of finite lifetime. By lifetime of this bubble, we mean the time interval during which the bubble is created, grows, collapses, and eventually disappears. Our group has been able to measure this lifetime as function of temperature. We have shown that it undergoes a dramatic transition while crossing the λ point[38] as shown in Figure 4a).

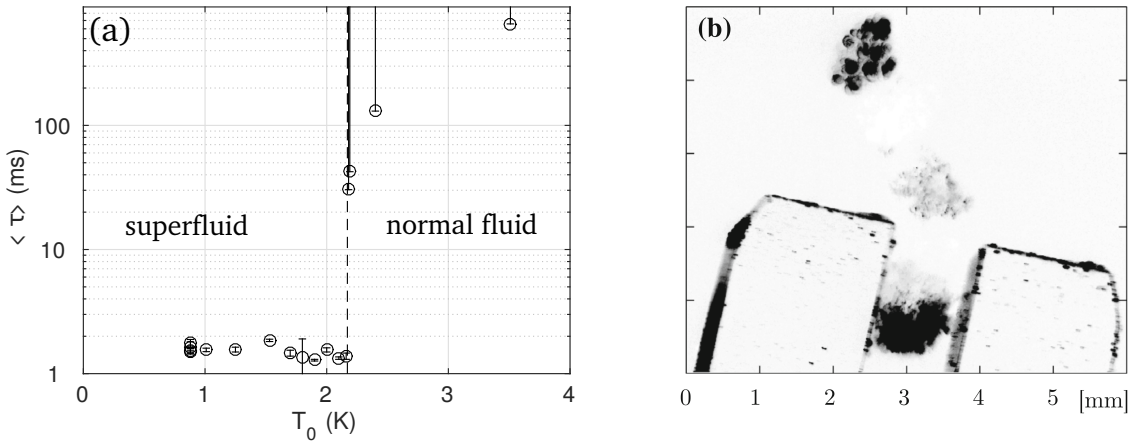

Figure 4: a) The lifetime of cavitation bubbles in liquid ^{4}He as a function of temperature as measured by the LKB's group in Paris, reprinted from [38]. b) The vibrating quartz tuning fork of the Prague's group produce heterogeneous cavitation bubbles in liquid He I, reprinted from [39]. Their lifetime is so long that the bubbles have time to leave their nucleation sites (between the prongs of the fork) and rise up because of buoyancy. Such behavior is never observed in He II in which bubbles collapse where they were created.

The Prague's group has also reported notably longer lifetimes of cavitation bubbles in He I than in He II[39].

The dynamics of the bubble collapse and hence the bubble lifetime is mainly governed by the way heat from the condensation of the gas is extracted away from the surface. In the superfluid case, heat is very efficiently transported by the propagation of a second sound wave [40] making the bubble lifetime much shorter than in the normal fluid where the latent heat of condensation must be evacuated via normal heat diffusion.

2.4 Overpressurised Liquid ^{4}He

The acoustic technique has been used in liquid ^{4}He not only to achieve negative pressure metastable states but also to overpressurize the liquid to metastable states with respect to the solid phase. Regarding Figure 1, the starting point of the experiment would be B with phase 2 being the liquid phase and phase 1 being the solid phase. The overpressurised metastable states are thus obtained during the positive swings of the wave. These overpressurised metastable states are interesting to study especially to check whether an intrinsic instability of the liquid→solid transition of liquid ^{4}He does exist. Indeed, in the case of liquid→gas transition, the spinodal limit of the metastable liquid at negative pressures is defined as the line of the Pressure/Temperature phase diagram where the speed of sound becomes zero. But this condition is not relevant for overpressurised liquid ^{4}He and hence a stability limit if it exists must invoke some other mechanism. It has been suggested that the "roton gap"can vanish in a high density (pressure) metastable superfluid state[41, 42]. If this happens, the liquid would be unstable against finite wavelength density fluctuations at the finite roton wavelength. This is expected to result in the spontaneous organization of the liquid in a periodic arrangement at this particular wavelength which is likely to evolve in the crystalline state[3]. The reality of this interesting scenario is still under debate. Numerics based on Quantum Monte Carlo Simulation at zero temperature performed by L. Vranješ *et al.*[43] have computed the roton gap energy Δ_R up to 275 bar where it is still about 3 K (the same numerics give 8.5 K at 0 bar). Although the error bar on the numerical data is pretty large, the authors fit their data with a decreasing exponential function suggesting Δ_R is never becoming zero. M. Rossi *et al.* have used a shadow path integral ground state (SPIGS) method to compute Δ_R between 0 and 89 atm and found a very different behavior as their calculation shows a linear dependence of Δ_R on pressure. A linear extrapolation of their results gives that Δ_R becomes zero for $P \sim 170$ bar[44]. It is interesting to note that the results for Δ_R of references [43] and [44] agree for pressures below 40 bar and match neutron scattering experimental measurements performed in porous Gelsil filled with liquid ^{4}He[3]. At much higher pressure however the two results deviate from each other and the much smaller numerical error bars reported by M. Rossi *et al.* than by the work [43] tend to favor the linear dependence of Δ_R on the liquid pressure.

Experiments using the converging acoustic wave technique have been conducted to overpressurize bulk liquid ^{4}He in the Balibar/Caupin's group. In a first paper, Werner *at al.* used a single hemispherical transducer resonating at about 1 MHz (thickness mode) to look for acoustic nucleation of ^{4}He crystal. They claimed that they had pressurized metastable liquid ^{4}He up to +160 bar without detecting crystal nucleation[45]. In these experiments, the authors were seeing nucleation events which they attribute to cavitation (nucleation of bubbles at negative pressures) even if the static pressure of the cell (point B of Fig. 1) was close to the liquid/solid coexistence line (25 bar). However, the Balibar/Caupin's group has reconsidered this conclusion after having conducted another experiment using a spherical acoustic transducer resonating at about 140 kHz (breathing mode). In this experimental work[46, 47], the authors were able to distinguish clearly in time nucleation events due to cavitation (gas bubbles) or nucleation of crystals that they do observe. However, no estimation of the pressure at which acoustic crystallization takes place is given. The authors argue that the way they measure cavitation pressures[4] could not be applied to the case of crystal nucleation probably because of non well understood non linearities in the sound propagation. Whether crystals nucleate at about 60 bar which is a numerical estimation the homogeneous nucleation pressure within the thin wall approximation of the standard nucleation theory[48][5] or at much higher pressures (170-300 bar) due to the vanishing of the roton gap is still an open question.

3 Metastable Solid ^{4}He

3.1 Experiment

An interesting possibility regarding the non-equilibrium phases of condensed ^{4}He is to apply the acoustic technique to the solid phase of ^{4}He in order to produce a low density (metastable) state of hcp solid ^{4}He. Indeed, at low temperatures ($0 < T < 1.4$ K), the melting line of ^{4}He is nearly horizontal in the T-P plane, in contrast with other simple solids where it is nearly vertical. In this unique situation, pressure is the efficient control parameter to explore the metastability region below the melting line. Metastable expanded solid helium-4 has also been considered in the past in view of achieving the Andreev-Lifshitz-Thouless scenario for supersolidity[49, 50, 51] by lowering the vacancy energy[52]. Finally, due to its extreme purity, it is an ideal candidate for studying homogeneous nucleation in solids.

[3] The roton wavelength is close to the average distance between ^{4}He atoms in the liquid

[4] namely the extrapolation of the $(P_{stat}, \rho_{stat} \cdot V_C)$ to $V_C = 0$, P_{stat}, ρ_{stat} are respectively the static pressure and densities of the cell and V_C the voltage needed to nucleate a crystal, as explained in section 2.1 for cavitation pressure measurements.

[5] As mentioned by Caupin and Maris in that paper, the thin wall approximation becomes "suspect"at such high pressure where the critical radius of a solid germ is comparable to the interatomic spacing.

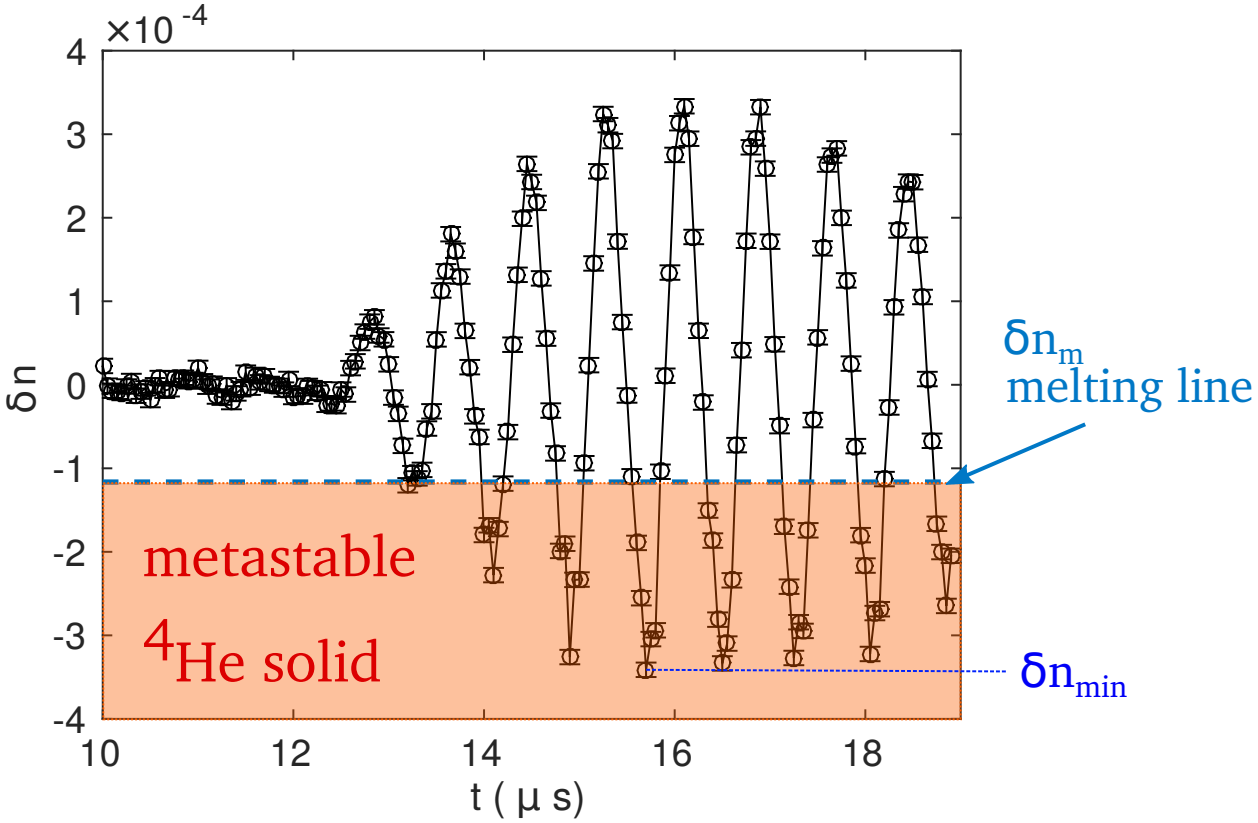

Figure 5: Time evolution of the refractive index variations at acoustic focus induced by a converging elastic wave in solid hcp ^{4}He at 1.1 K. The crystal is initially set at a pressure that corresponds to a density higher than that of the melting curve (point A of Fig. 1). The corresponding optical refractive index variations between point A and the melting curve is given by δn_m (see text). During the negative swing of the acoustic wave, the local density is below the static melting line, meaning that hcp solid ^{4}He is in a metastable state. δn_{min} is the maximum of the negative refractive index variations (point B of Fig. 1) whose value depends on the driving voltage of the piezo transducer.

The principle of the experiment aiming at producing hcp solid ^{4}He using the acoustic technique is again given by Figure 1 with phase 1 being the (stable) solid phase and phase 2 the (stable) superfluid phase. Solid ^{4}He at point B of Figure 1 is in a metastable state. Compared to the liquid case, a difficulty arises from the anisotropy of the sound propagation in hexagonal close-packed (hcp) solid helium-4. The wavesurface of a pressure/density wave front originating from a point source is not a sphere but has an ovoid shape extended along the radial symmetry axis of the crystal ("c-axis"). Hence hemispherical transducers cannot focus sound waves in that case. In a hcp lattice, three modes of elastic waves can propagate, a pure transverse wave (shear wave) often labeled T_1, a quasi-transverse mode labeled T_2 and a quasi-longitudinal wave labeled L. Crepeau *et al.* have measured sound velocities in hcp solid ^{4}He crystals of known orientation[53]. From these measurements, they can deduce velocity surfaces and wave surfaces in hcp solid ^{4}He for the three acoustic modes. The L-wave is the mode which gives the greatest variation of the density for a given displacement of the transducer[54]. In our group, we have designed a piezo electric transducer shaped to the wave surface of the L-mode[55] in order to produce metastable hcp solid ^{4}He. Using the same optical set-up than the one sketched in Figure 3, we have been able to measure hcp solid ^{4}He at densities below the melting line for temperatures ranging between 1 and 1.4 K. Figure 5 shows the time evolution of the refractive index variations at acoustic focus induced by the converging elastic wave in solid hcp ^{4}He. On this peculiar example, the crystal was initially set at the point $A(T_0 = 1.13\ \text{K}, P_0 = 26.36\ \text{bar})$ and the melting pressure is $P_m(T_0) = 25.49$ bar. Knowing the values of $P_m(T_0)$ and P_0 and the Equation of State of stable hcp solid ^{4}He[6], the melting line can be placed in this Figure. One can see that during the negative swing of the acoustic wave, the local density is clearly below the static melting line, meaning that metastable hcp solid ^{4}He has indeed been produced and observed[57].

When the driving voltage of the PZT exceeds a threshold value, it was observed that crystal breaks in the vicinity of acoustic focus. An extended defect with a different density appears near the focal point. Such a defect is shown in the inset of Figure 6. It should be noted that this optical defect displays a net and sharp interface with the surrounding solid hcp ^{4}He monocrystal. This suggests that it corresponds to another phase than hcp solid ^{4}He rather than a disordered hcp ^{4}He crystal. Assuming that the EOS of stable solid ^{4}He holds in the metastable domain, it was found that the pressure at which the instability occurs lies between 3 and 4 bar below the melting line and is not really temperature dependent as it can be seen in Figure 6.

[6]Grilly [56] has measured that the quasi isotropic compressibility $\chi = 269$ bar of hcp solid ^{4}He is relatively independent (maximum of 2% variations) of pressure for pressures between 25 and 27 bar on the melting curve.

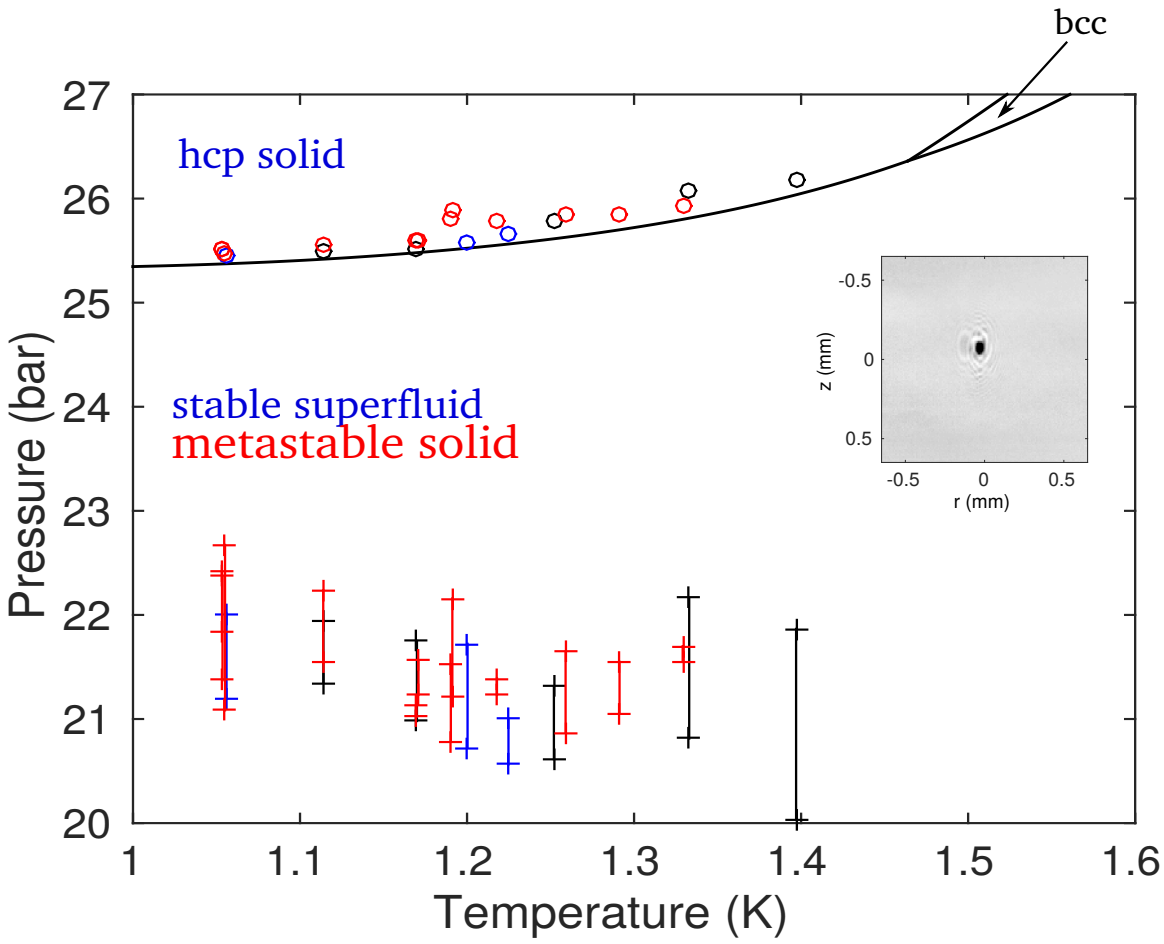

Figure 6: Phase diagram of ^{4}He summarizing the experimental results concerning the destabilization of metastable solid helium obtained by Souris *et al.*[57]. The color circles represent the starting point of each acoustic experiment (point A of Fig. 1) and the color corresponding bar in the metastable domain is an interval of the estimated pressure value at which the instability occurs. Inset : Raw interferometric image of the optical defect corresponding to the destabilization of metastable solid ^{4}He.

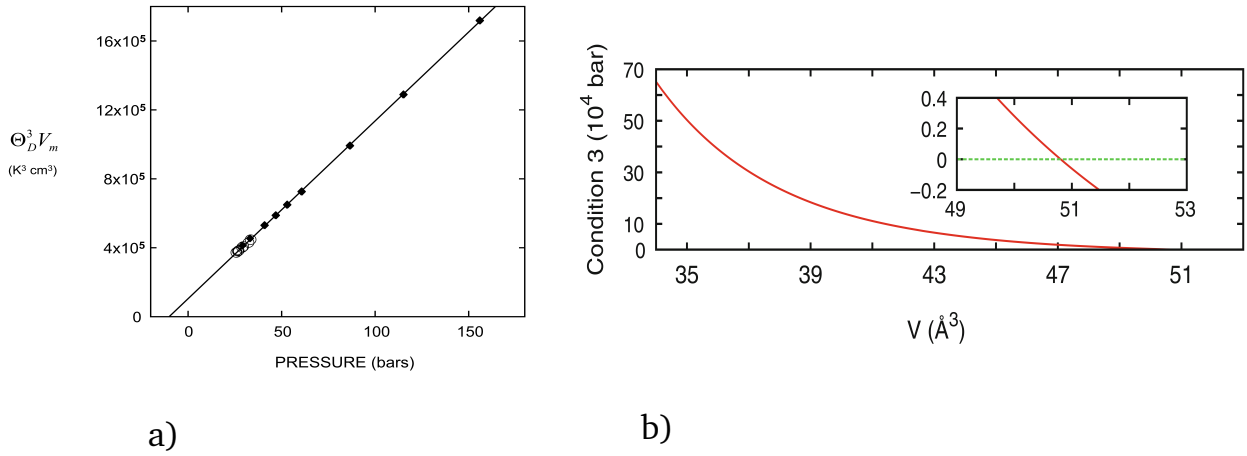

Figure 7: a) H. Maris estimates the spinodal pressure of hcp solid ^{4}He by fitting available data of the Debye temperature using equation (2). He finds the spinodal pressure to be $P_c \sim -10$ bar. Reprinted from[58]. b) Cazorla and Boronat computation of condition C[3] (see text and equation (3) for the stability limit of hcp solid ^{4}He. Reprinted from [61].

3.2 Theory

Few theoretical works have been devoted to the properties of metastable solid ^{4}He below the normal melting pressure.

Spinodal pressure of hcp solid ^{4}He. H. Maris attempted to apply to metastable solid ^{4}He[58] the same ideas that he had apply to metastable liquid ^{4}He[24]. His calculation of the spinodal pressure of liquid ^{4}He was made by fitting the sound velocity pressure dependence by a cubic law $c^3 = b(P - P_c)$, the spinodal pressure being the pressure at which the sound velocity vanishes. In the case of hcp solid ^{4}He, estimating its stability limit is more complicated because of two reasons. The first one is that metastable solid ^{4}He can destabilize not because of the vanishing of sound velocity but due to creation and proliferation of crystalline defects such as vacancies or dislocations. Forgetting this issue for now, another complication is that, as already mentioned, three different sound modes can propagate in hcp solid ^{4}He, a quasi-longitudinal one (L) and two quasi transverse modes (T_1 and T_2) and their propagation velocity depends on the propagation direction[53]. If linked to the vanishing of sound velocity, the stability limit of the the crystal should then be given by the condition that one of acoustic mode velocity in any direction becomes zero. As there are few available data on the dependence to pressure (molar volume) of sound velocities in hcp solid ^{4}He, Maris used the Debye temperature to estimate the spinodal limit of the crystal. The Debye temperature is given by :

$$\Theta_D = \frac{\hbar v_D}{k} \left(\frac{6\pi^2 N_A}{V_m} \right)^{1/3} \tag{1}$$

where $\hbar$ and k_B are the reduced Planck and Boltzmann constants, N_A the Avogadro number, V_m the molar volume of the solid and v_D the Debye velocity which is linked to the angular average velocities of the acoustic longitudinal and transverse modes. The Debye temperature of stable hcp solid ^{4}He has been accurately measured over a wide range of pressure[59, 60]. As shown in Figure 7, Maris found that the quantity $V_m \Theta_D^3$ can be very well fitted by the relation :

$$V_m \Theta_D^3 = b(P - P_c) \tag{2}$$

where b is a constant, P the pressure of the solid and P_c the critical pressure at which the quantity $V_m \Theta_D^3$ becomes zero. If one assumes that P_c corresponds to the spinodal pressure of hcp ^{4}He, the fit of Figure 7 gives a spinodal pressure of about -10 bar.

An other estimate of the spinodal pressure of hcp solid ^{4}He has been given by Cazorla and Boronat[61]. The hcp lattice is described by five elastic constants which are usually labeled C_{11}, C_{12}, C_{13}, C_{33} and C_{44} according to Voigt notations. It has been shown that to be mechanically stable, an hcp crystal subject to an isotropic external pressure P must fulfill the following conditions[62]:

$$\begin{aligned} C_{44} - P &> 0 \ \text{[C1]} \\ C_{11} - C_{12} - 2P &> 0 \ \text{[C2]} \\ (C_{33} - P)(C_{11} + C_{12}) - 2(C_{13} + P)^2 &> 0 \ \text{[C3]} \end{aligned} \tag{3}$$

Cazorla and Boronat have used a Diffusion Monte Carlo method to compute the elastic constants of hcp solid ^{4}He as a function of the volume of the unit cell V and hence of an isotropic external pressure P. Their results shown in Figure 7 b) indicate that the here-above [C3] condition is the first of the three not to be satisfied as V increases for value $V_c = 50.81(5)$ Å^3 corresponding to a critical pressure $P_s = -33.8(1)$ bar. This result is very different from the Maris estimation (~ -10 bar). The discrepancy between the two results originates from the fact that, as mentioned by Cazorla and Boronat, the computation of the C_{ij} in metastable hcp solid ^{4}He leads to a non vanishing value of the Debye temperature at $P \sim -10$ bar and for all pressures down and even at the critical pressure P_s (violation of condition [C3]).

In any case, these numbers are far away from the experimental value of the destabilization pressure of hcp solid ^{4}He mentioned in the previous sections which is about +21 bar. However, at a given temperature $T \neq 0$ the spinodal limit does not correspond to the nucleation line. Maris estimated the nucleation pressure of a bubble of liquid in solid hcp ^{4}He in the framework of standard nucleation theory[58]. At T=1 K, he founds that the nucleation probability is not negligible only for pressure close to -10 bar, which is his estimation of the solid spinodal pressure.

3.3 On the possible role of crystalline defects.

The large gap between theoretical estimates and experimental destabilization pressure of hcp solid ^{4}He suggests that other mechanisms than bulk homogeneous nucleation should be considered to explain these observations.

As shown in Figure 8, an experiment performed by our group on metastable hcp solid ^{4}He has shown that the exact time of the solid destabilization corresponds indeed to negative pressure swings of the acoustic wave, when the solid is exploring its metastable state [63]. This was of course expected. Less expected was the fact that, for the seven experiments performed at the lowest driving voltage ($V_{PZT} = 57$ V), the instability systematically occurs during the oscillation *after* the one of minimum pressure. If a simple nucleation threshold did exist as homogeneous nucleation theory does suggest, the 57 V instabilities would have nucleated at least one oscillation before the one observed. Even if we assume that there is only some probability of breaking during a certain swing, as soon as the amplitude is large enough, this result doesn't seem compatible with a simple threshold. Indeed, the two preceding oscillations have amplitudes equal to or larger than the one where events are observed. Attributing equal probabilities for a nucleation event in each of these oscillations, the probability for 7 events lying in the last oscillation would be only $(1/3)^7 \simeq 4 \cdot 10^{-4}$.

This suggests that fatigue effects could play an important role in triggering the instability. Fatigue is the weakening of a material caused by repeatedly applied stresses leading to the creation and proliferation of crystalline defects inside the sample.

Vacancy proliferation A possibility is that solid ^{4}He in its metastable state makes easier the creation and proliferation of vacancies. Pollet *et al.* have computed the density dependence of the vacancy and interstitial chemical potentials in hcp stable solid ^{4}He using a worm algorithm path integral Monte Carlo method[64]. The authors used the near perfect linear density dependence of the chemical potentials to extrapolate their values in the metastable domain assuming an isotropic (metastable) pressure applied to solid. At a critical density n_c, the creation of a pair vacancy/interstitial costs zero energy and hence the solid must become unstable. Assuming a constant compressibility of the solid in the metastable domain, the destabilization pressure P_c corresponding to n_c is around 0 bar. This number is still far from the experimental estimate for the destabilization of the solid (+21bar, [57])

Dislocation proliferation. Dislocations are other crystalline defects that can weaken the solid. Cheng and Beamish [65] have recently shown that sheared solid ^{4}He shows clear evidence of plastic flow. This plastic flow is the signature of the proliferation of dislocations into the solid. It is then possible that, in the experiments producing metastable hcp solid ^{4}He where comparable strains to that of Cheng and Beamish are produced (~ 0.2 %), dislocations proliferate[7]. Unfortunately, there is for now no available nucleation theory of solid ^{4}He assisted by dislocations. This would be of great interest in order to better understand the observed destabilization threshold of metastable hcp solid ^{4}He.

[7]It must be noted however that in the Cheng and Beamish experiment[65], the strain rates are about 10^{-5} s^{-1}, far away from the MHz modulation of ref [57].

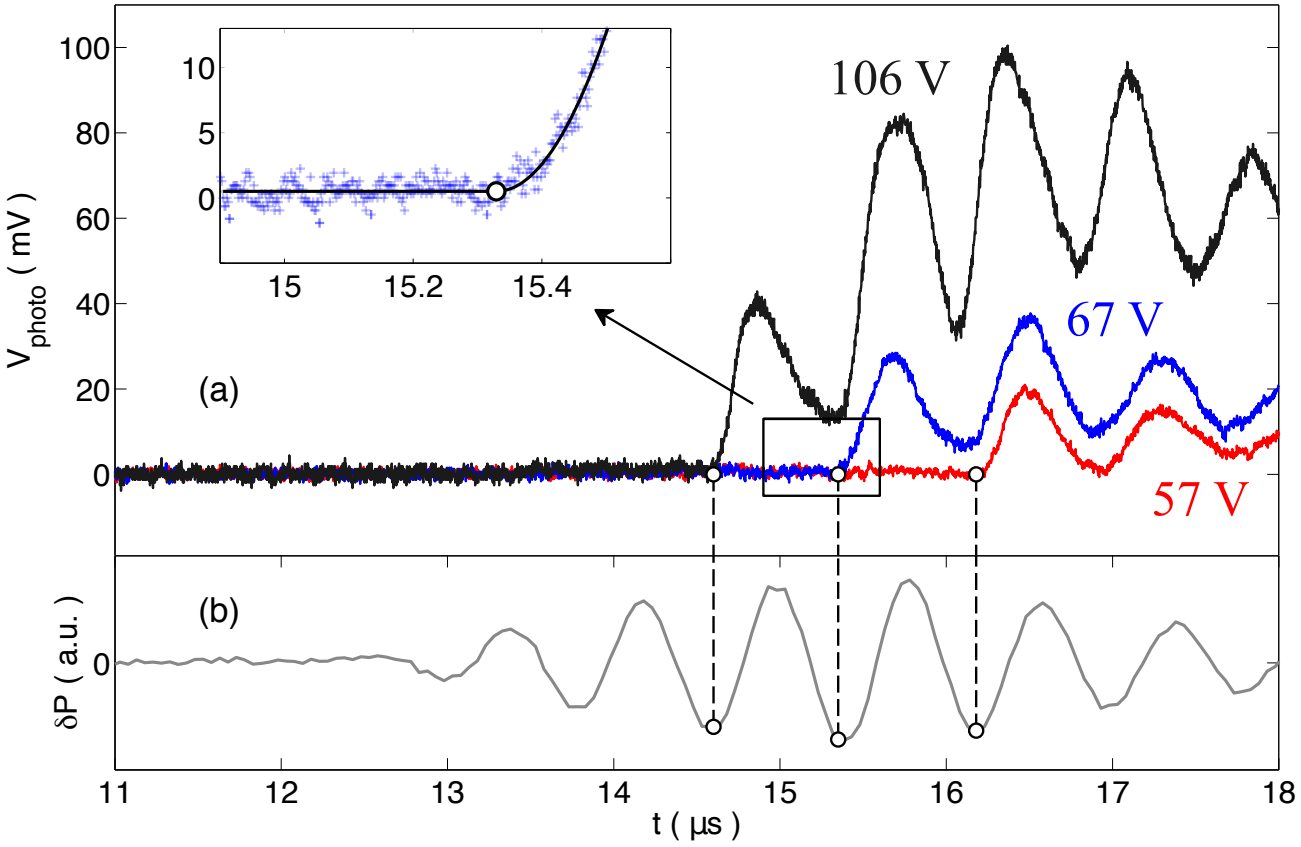

Figure 8: a) Photodiode voltage proportional to the light scattered by an optical defect (see inset of Fig. 6) created in metastable hcp solid ^{4}He as function of time for 3 experiments creating solid instabilities at different PZT driving voltages. The sharp increase of the signal corresponds to the appearance of the instability. The open circles give the time of birth t_0. Inset: closer look at the creation time of the instability. b) Pressure variations at acoustic focus in arbitrary units for temporal reference. Reprinted from [63]

4 Conclusion and open problems

^{4}He is certainly a model system to study metastable states of condensed matter, spinodal limits and nucleation theories. This is mainly because the atom interactions are weak and impurities almost inexistent. However, even in such a "simple "system, nucleation mechanisms of a stable phase in the metastable one are still nowadays not completely understood.

Regarding metastable liquid ^{4}He and cavitation in it, recent measurements of the cavitation threshold have re-opened the debate on heterogeneous nucleation in this perfectly wetting system. Unfortunately, no theory is available to understand clearly what could be happening in experiments showing evidences of heterogeneous cavitation. New ideas, calculations and other experiments are needed to solve this interesting problem. Recent experiments looking at the typical lifetime of cavitation bubbles in He I and He II showing a dramatic difference in both cases have also been presented. Finally for metastable liquid ^{4}He, it has been pointed out that overpressurised (high density) metastable states of the liquid have been experimentally produced and crystallization in them observed. Whether this crystallization is linked to the vanishing of the roton gap in the metastable state is still at present an open question.

Regarding metastable phases of solid ^{4}He, metastable hcp solid ^{4}He has been newly experimentally produced and observed. An unexpected instability of the metastable solid phase was found at pressures which are not compatible with theoretical estimates of the spinodal limit of the solid phase. Here also, the possibility of heterogeneous nucleation of the liquid phase on crystalline defects of the metastable solid is an open and exciting question for both experimentalists and theoreticians.

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References

[1] F. Albergamo, J. Bossy, P. Averbuch, H. Schober, H.R. Glyde, Phys. Rev. Lett. **92**, 235301 (2004). DOI 10.1103/PhysRevLett.92.235301. URL https://link.aps.org/doi/10.1103/PhysRevLett.92.235301

[2] M.S. Bryan, P.E. Sokol, Phys. Rev. B **97**, 184511 (2018). DOI 10.1103/PhysRevB.97.184511. URL https://link.aps.org/doi/10.1103/PhysRevB.97.184511

[3] J.V. Pearce, J. Bossy, H. Schober, H.R. Glyde, D.R. Daughton, Mulders, N., Phys. Rev. Lett. **93**, 145303 (2004). DOI 10.1103/PhysRevLett.93.145303. URL https://link.aps.org/doi/10.1103/PhysRevLett.93.145303

[4] J.A. Nissen, E. Bodegom, L.C. Brodie, J.S. Semura, Phys. Rev. B **40**, 6617 (1989). DOI 10.1103/PhysRevB.40.6617. URL https://link.aps.org/doi/10.1103/PhysRevB.40.6617

[5] S. Balibar, Journal of Low Temperature Physics **129**(5), 363 (2002). DOI 10.1023/A:1021412529571. URL https://doi.org/10.1023/A:1021412529571

[6] D.N. Sinha, J.S. Semura, L.C. Brodie, Phys. Rev. A **26**, 1048 (1982). DOI 10.1103/PhysRevA.26.1048. URL https://link.aps.org/doi/10.1103/PhysRevA.26.1048

[7] N.M. Semenova, G.V. Ermakov, Journal of Low Temperature Physics **74**(1), 119 (1989). DOI 10.1007/BF00681755. URL https://doi.org/10.1007/BF00681755

[8] K. Nishgaki, Y. Saji, Journal of the Physical Society of Japan **52**(7), 2293 (1983). DOI 10.1143/JPSJ.52.2293. URL https://doi.org/10.1143/JPSJ.52.2293

[9] S.C. Hall, J. Classen, C.K. Su, H.J. Maris, Journal of Low Temperature Physics **101**(3), 793 (1995). DOI 10.1007/BF00753392. URL https://doi.org/10.1007/BF00753392

[10] M.S. Pettersen, S. Balibar, H.J. Maris, Phys. Rev. B **49**, 12062 (1994). DOI 10.1103/PhysRevB.49.12062. URL https://link.aps.org/doi/10.1103/PhysRevB.49.12062

[11] F. Caupin, S. Balibar, Phys. Rev. B **64**, 064507 (2001). DOI 10.1103/PhysRevB.64.064507. URL http://link.aps.org/doi/10.1103/PhysRevB.64.064507

[12] Appert, C., Tenaud, C., Chavanne, X., Balibar, S., Caupin, F., d'Humires, D., Eur. Phys. J. B **35**(4), 531 (2003). DOI 10.1140/epjb/e2003-00307-0. URL http://dx.doi.org/10.1140/epjb/e2003-00307-0

[13] A. Qu, A. Trimeche, J. Dupont-Roc, J. Grucker, P. Jacquier, Physical Review B **91**(21), 214115 (2015). URL https://journals.aps.org/prb/abstract/10.1103/PhysRevB.91.214115

[14] L. Landau, E. Lifshitz, *Statistical Physics, 3rd Edition, Chapter 162* (Elsevier / Butterworth - Heinemann, 2013)

[15] D.M. Jezek, M. Guilleumas, M. Pi, M. Barranco, J. Navarro, Phys. Rev. B **48**, 16582 (1993). DOI 10.1103/PhysRevB.48.16582. URL http://link.aps.org/doi/10.1103/PhysRevB.48.16582

[16] S.C. Hall, H.J. Maris, Journal of Low Temperature Physics **107**(3), 263 (1997). DOI 10.1007/BF02397457. URL https://doi.org/10.1007/BF02397457

[17] G.H. Bauer, D.M. Ceperley, N. Goldenfeld, Phys. Rev. B **61**, 9055 (2000). DOI 10.1103/PhysRevB.61.9055. URL http://link.aps.org/doi/10.1103/PhysRevB.61.9055

[18] A. Guirao, M. Centelles, M. Barranco, M. Pi, A. Polls, X. Vinas, Journal of Physics: Condensed Matter **4**(3), 667 (1992). DOI 10.1088/0953-8984/4/3/008. URL https://iopscience.iop.org/article/10.1088/0953-8984/4/3/008

[19] F. Souris, J. Grucker, J. Dupont-Roc, P. Jacquier, A. Arvengas, F. Caupin, Applied Optics **49**, 6127. URL https://www.osapublishing.org/ao/abstract.cfm?uri=ao-49-31-6127

[20] A. Qu, PhD Thesis, Universit Pierre et Marie Curie, Paris, France (2017). URL https://tel.archives-ouvertes.fr/tel-01591897

[21] H.J. Maris, J. Low Temp. Phys. **94**, 125. URL https://link.springer.com/article/10.1007/BF00755421

[22] F. Dalfovo, A. Lastri, L. Pricaupenko, S. Stringari, J. Treiner, Phys. Rev. B **52**, 1193 (1995). DOI 10.1103/PhysRevB.52.1193. URL http://link.aps.org/doi/10.1103/PhysRevB.52.1193

[23] J. Boronat, J. Casulleras, J. Navarro, Phys. Rev. B **50**, 3427 (1994). DOI 10.1103/PhysRevB.50.3427. URL http://link.aps.org/doi/10.1103/PhysRevB.50.3427

[24] H.J. Maris, D.O. Edwards, J. Low Temp. Phys. **129**, 1 (2002). URL https://link.springer.com/article/10.1023/A:1020060700534

[25] G.W. Faris, L.E. Jusinski, A.P. Hickman, J. Opt. Soc. Am. B **10**(4), 587 (1993). DOI 10.1364/JOSAB.10.000587. URL http://josab.osa.org/abstract.cfm?URI=josab-10-4-587

[26] H. Schubert, P. Leiderer, H. Kinder, Journal of Low Temperature Physics **39**(3), 363 (1980). DOI 10.1007/BF00115626. URL https://doi.org/10.1007/BF00115626

[27] H. Maris, Journal of Low Temperature Physics **98**(5-6), 403 (1995). DOI 10.1007/BF00752276. URL http://dx.doi.org/10.1007/BF00752276

[28] F. Dalfovo, Phys. Rev. B **46**, 5482 (1992). DOI 10.1103/PhysRevB.46.5482. URL https://link.aps.org/doi/10.1103/PhysRevB.46.5482

[29] W.H. Zurek, Nature **317**, 505 (1985). URL https://doi.org/10.1038/317505a0

[30] P.C. Hendry, N.S. Lawson, R.A.M. Lee, P.V.E. McClintock, C.D.H. Williams, Nature **368**(6469), 315 (1994). DOI 10.1038/368315a0. URL https://doi.org/10.1038/368315a0

[31] M.E. Dodd, P.C. Hendry, N.S. Lawson, P.V.E. McClintock, C.D.H. Williams, Phys. Rev. Lett. **81**, 3703 (1998). DOI 10.1103/PhysRevLett.81.3703. URL https://link.aps.org/doi/10.1103/PhysRevLett.81.3703

[32] V.B. Efimov, O.J. Griffiths, P.C. Hendry, G.V. Kolmakov, P.V.E. McClintock, L. Skrbek, Phys. Rev. E **74**, 056305 (2006). DOI 10.1103/PhysRevE.74.056305. URL https://link.aps.org/doi/10.1103/PhysRevE.74.056305

[33] A. Ganshin, M. Mohazzab, N. Mulders, Journal of Low Temperature Physics **134**(1), 477 (2004). DOI 10.1023/B:JOLT.0000012598.22534.c3. URL https://doi.org/10.1023/B:JOLT.0000012598.22534.c3

[34] P.J. Nacher, J. Dupont-Roc, Phys. Rev. Lett. **67**, 2966 (1991). DOI 10.1103/PhysRevLett.67.2966. URL https://link.aps.org/doi/10.1103/PhysRevLett.67.2966

[35] X. Chavanne, S. Balibar, F. Caupin, C. Appert, D. d'Humières, Journal of low temperature physics **126**(1-2), 643 (2002)

[36] M. Blakov, D. Schmoranzer, L. Skrbek, Low Temperature Physics **34**(4), 298 (2008). DOI 10.1063/1.2908890. URL https://doi.org/10.1063/1.2908890

[37] M. Blažková, T.V. Chagovets, M. Rotter, D. Schmoranzer, L. Skrbek, Journal of Low Temperature Physics **150**(3), 194 (2008). DOI 10.1007/s10909-007-9533-4. URL https://doi.org/10.1007/s10909-007-9533-4

[38] A. Qu, A. Trimeche, P. Jacquier, J. Grucker, Physical Review B **93**(17), 174521 (2016). URL https://journals.aps.org/prb/abstract/10.1103/PhysRevB.93.174521

[39] D. Duda, P. Švančara, M. La Mantia, M. Rotter, D. Schmoranzer, O. Kolosov, L. Skrbek, Journal of Low Temperature Physics **187**(5), 376 (2017). DOI 10.1007/s10909-016-1684-8. URL https://doi.org/10.1007/s10909-016-1684-8

[40] J. Wilks, *The properties of liquid and solid helium* (Oxford University Press, 1967)

[41] T. Schneider, C.P. Enz, Phys. Rev. Lett. **27**, 1186 (1971). DOI 10.1103/PhysRevLett.27.1186. URL https://link.aps.org/doi/10.1103/PhysRevLett.27.1186

[42] P. Nozières, Journal of Low Temperature Physics **142**(1), 91 (2006). DOI 10.1007/s10909-005-9413-8. URL https://doi.org/10.1007/s10909-005-9413-8

[43] L. Vranješ, J. Boronat, J. Casulleras, C. Cazorla, Phys. Rev. Lett. **95**, 145302 (2005). DOI 10.1103/PhysRevLett.95.145302. URL https://link.aps.org/doi/10.1103/PhysRevLett.95.145302

[44] M. Rossi, E. Vitali, L. Reatto, D.E. Galli, Phys. Rev. B **85**, 014525 (2012). DOI 10.1103/PhysRevB.85.014525. URL https://link.aps.org/doi/10.1103/PhysRevB.85.014525

[45] F. Werner, G. Beaume, A. Hobeika, S. Nascimbène, C. Herrmann, F. Caupin, S. Balibar, Journal of Low Temperature Physics **136**(1), 93 (2004). DOI 10.1023/B:JOLT.0000035372.69378.db. URL https://doi.org/10.1023/B:JOLT.0000035372.69378.db

[46] R. Ishiguro, F. Caupin, S. Balibar, EPL (Europhysics Letters) **75**(1), 91 (2006)

[47] R. Ishiguro, F. Caupin, S. Balibar, Journal of Low Temperature Physics **148**(5), 645 (2007). DOI 10.1007/s10909-007-9477-8. URL https://doi.org/10.1007/s10909-007-9477-8

[48] H.J. Maris, F. Caupin, Journal of Low Temperature Physics **131**(1), 145 (2003). DOI 10.1023/A:1022813514979. URL https://doi.org/10.1023/A:1022813514979

[49] A.F. Andreev, I.M. Lifshitz, Sov. Phys-JETP **29**, 1107 (1969). URL http://www.jetp.ac.ru/cgi-bin/e/index/e/29/6/p1107?a=list

[50] D.J. Thouless, Ann. Phys. **52**, 403 (1969)

[51] S. Balibar, Nature **464**, 176 (2010). URL https://www.nature.com/articles/nature08913

[52] L. Pollet, M. Boninsegni, A.B. Kuklov, N.V. Prokof'ev, B.V. Svistunov, M. Troyer, Phys. Rev. Lett. **101**(9), 097202 (2008). DOI 10.1103/PhysRevLett.101.097202

[53] R. Crepeau, D. Lee, Phys. Rev. A **6**, 516. URL https://link.aps.org/doi/10.1103/PhysRevA.6.516

[54] F. Souris, PhD Thesis, Universit Pierre et Marie Curie, Paris, France (2013). URL https://tel.archives-ouvertes.fr/tel-00942738

[55] F. Souris, J. Grucker, N. Garroum, A. Leclercq, J.M. Isac, J. Dupont-Roc, P. Jacquier, Review of Scientific Instruments **85**(6), 064902 (2014). DOI 10.1063/1.4881535. URL https://doi.org/10.1063/1.4881535

[56] E.R. Grilly, Journal of Low Temperature Physics **11**(1), 33 (1973). DOI 10.1007/BF00655035. URL https://doi.org/10.1007/BF00655035

[57] F. Souris, J. Grucker, J. Dupont-Roc, P. Jacquier, EPL (Europhysics Letters) **95**(6), 66001 (2011). URL https://epljournal.edpsciences.org/articles/epl/abs/2011/18/epl13806/epl13806.html

[58] H.J. Maris, Journal of Low Temperature Physics **155**(5), 290 (2009). DOI 10.1007/s10909-009-9881-3. URL https://doi.org/10.1007/s10909-009-9881-3

[59] G. Ahlers, Phys. Rev. A **2**, 1505 (1970). DOI 10.1103/PhysRevA.2.1505. URL https://link.aps.org/doi/10.1103/PhysRevA.2.1505

[60] W.R. Gardner, J.K. Hoffer, N.E. Phillips, Phys. Rev. A **7**, 1029 (1973). DOI 10.1103/PhysRevA.7.1029. URL https://link.aps.org/doi/10.1103/PhysRevA.7.1029

[61] C. Cazorla, J. Boronat, Journal of Low Temperature Physics **180**(1), 20 (2015). DOI 10.1007/s10909-014-1238-x. URL https://doi.org/10.1007/s10909-014-1238-x

[62] G.V. Sin'ko, N.A. Smirnov, Journal of Physics: Condensed Matter **14**(29), 6989 (2002). DOI 10.1088/0953-8984/14/29/301. URL https://iopscience.iop.org/article/10.1088/0953-8984/14/29/301/meta

[63] F. Souris, A. Qu, J. Dupont-Roc, J. Grucker, P. Jacquier, Journal of Low Temperature Physics **179**(5-6), 390 (2015). URL https://link.springer.com/article/10.1007/s10909-013-1037-9

[64] M. Boninsegni, N. Prokof'ev, B. Svistunov, Phys. Rev. Lett. **96**, 070601 (2006). DOI 10.1103/PhysRevLett.96.070601. URL https://link.aps.org/doi/10.1103/PhysRevLett.96.070601

[65] Z.G. Cheng, J. Beamish, Phys. Rev. Lett. **121**, 055301 (2018). DOI 10.1103/PhysRevLett.121.055301. URL https://link.aps.org/doi/10.1103/PhysRevLett.121.055301