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TOPICAL REVIEW

Supersolidity and disorder

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Abstract

A solid is called 'supersolid' if it exhibits superfluid properties. Supersolidity is a paradoxical phenomenon whose understanding has become a major challenge since 2004, when Kim and Chan first observed what could be mass superflow through solid helium 4. In this review, we describe how successive experiments indicated that what was observed in helium 4 was not intrinsic properties of the crystalline state as originally proposed 35 years before. Disorder coming from how the solid is grown (dislocations, grain boundaries and other interfaces, liquid or glassy regions, impurities...) was shown to play an essential role. However, one does not know yet which type of disorder is involved or by which mechanism it leads to the observed properties. Furthermore, all the experimental features probably cannot be explained by a common mechanism. Recent measurements of the shear modulus of helium 4 crystals could even be explained without the need of any superfluidity. In fact, many theoretical predictions need to be checked experimentally, so the whole issue is far from understood. Even some crucial experiments would need to be repeated more systematically. The present review of the experimental observations and theoretical scenarios raises a series of questions which call for answers.

(Some figures in this article are in colour only in the electronic version)

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1. Introduction

For a physical system, to be solid and superfluid at the same time, that is 'supersolid', is obviously paradoxical. The usual definition of a solid is a state with non-zero elastic shear modulus. One should probably specify 'at low frequency and

small stress'. The non-zero shear modulus is a consequence of the localization of atoms, which breaks the translation symmetry of the system. On the contrary, in a superfluid, atoms are indistinguishable and delocalized, and mass can flow without resistance; this is the case for *liquid* helium 4 below around 2 K [1, 2]. Still, it was proposed in 1969–70 that a quantum crystal with large quantum fluctuations of atoms around the lattice sites could be supersolid, in particular if it contained delocalized vacancies in the zero temperature limit [3–7].

Crystals are examples of solids where the translation symmetry is spontaneously broken by the appearance, at the crystallization point, of mass density waves with well defined wavevectors (the Bragg vectors). The possible existence of supersolidity leads to the question: is it possible that a system is periodic in space without being localized? For this, long range atom exchange is necessary. But is this exchange possible in perfect crystals or does it need the presence of vacancies or various kinds of defects? This is a fundamental problem which is at the heart of the whole issue of supersolidity.

There were early unsuccessful searches for a supersolid state [8, 9]. The first sign of a possible transition came from the experiments done by Kim and Chan in 2004. They discovered surprising anomalies in the behaviour of a torsional oscillator (TO) filled with solid helium¹ and presented them as possible experimental evidence for the supersolidity of solid helium [10, 11]. Since 2004, supersolidity has become controversial and fascinating. Despite the efforts of many experimental and theoretical groups, the whole issue is still unsettled at the end of the year 2007. In his 2007 article, Prokof'ev [12] has reviewed most of the theoretical aspects of supersolidity. In the present review, we have put more emphasis on experiments and tried to discriminate between various ideas and models. As we shall see, the involved community is far from having reached a general consensus on the interpretation of all observations, but it should be useful to summarize what has been understood and what should now be considered. This is the goal of this review, which is organized as follows.

In section 2, we describe the first TO experiments. In section 3, we consider the possible explanation in terms of superfluid vacancies. In section 4, we consider disorder in solid helium samples: dislocations, grain boundaries, glassy or liquid regions in inhomogeneous samples and helium 3 impurities. In section 5, we review a number of measurements of mechanical properties and their possible interpretations. We present in section 6 a few additional remarks on quantum gases, hysteresis, critical velocities, neutron and x-ray scattering measurements, pressure dependence, hydrogen, etc. Finally, in section 7, we draw a few conclusions and summarize questions which call for answers.



Figure 1. The TO used by Kim and Chan. The torsion rod is twisted by the sample cell oscillation, driven and detected by the side electrodes.

2. First experimental observations

2.1. Principle of the torsional oscillator experiment

The idea to use the change in resonance frequency of a TO as a measure of the superfluid fraction goes back to Andronikashvili [13], who used it with bulk liquid helium 4. The technique was later refined by using a TO with a high quality factor to measure the superfluid fraction in helium 3 [14, 15]. Since then, it has been widely used, in particular to study helium films or liquid helium confined in porous materials.

Let us first describe the set-up which was used by Kim and Chan [10] and is shown in figure 1. The torsion cell is a box suspended by its fill line, which is a vertical hollow rod. Two planar electrodes are attached on the sides of the torsion cell. They are coupled capacitively to two other electrodes: the detection electrode produces an ac voltage when the cell oscillates, and, thanks to a lock-in amplifier, the drive electrode keeps the oscillation in resonance. Because of the high quality factor of the torsion cell (typically 10⁶), the resonance period τ (typically 1 ms) can be measured with high accuracy. As a reference, τ is measured as a function of temperature for the empty cell:

$$\tau_0 = 2\pi \sqrt{\frac{I}{G}} \tag{1}$$

where I is the inertia momentum of the torsion bob, and G the torsion spring constant, determined by the rod shape and material. When the cell is filled with helium at high temperature, the added mass increases I and thus τ . In the

¹ From now on in this review, helium means helium 4 except when there is a possible ambiguity because, for example, we have to consider the effect of helium 3 impurities.



Figure 2. (A) The period shift (left scale, filled symbols) and the relative amplitude (right scale, open symbols) of the TO for different maximum oscillation velocities (see legend) as measured by Kim and Chan [10] in an annular cell at 51 bar. (B) The same measurements with a blocked annular channel at 36 bar and with a velocity of $3 \ \mu m \ s^{-1}$ (open stars, relative amplitude; filled stars, period shift of the filled cell; crosses, period shift of the empty cell). The period shift is measured with respect to the extrapolation to T = 0 of the empty cell (dotted line). When a barrier is placed across the helium cell (B), the period shift is strongly reduced with respect to the unblocked cell (A) and no attenuation peak can be seen.

case of *liquid* helium 4, as the cell is cooled down below a critical temperature T_0 , where superfluidity appears, the mass coupled to the box decreases, consequently *I* and the period τ . Due to the existence of a critical velocity in the hydrodynamics of superfluids, the observed change in inertia is largest in the limit of small drive amplitude. This is called 'non-classical rotational inertia' (NCRI). The relative change in the inertia momentum is called the NCRI fraction (NCRIF) and it is identified with the superfluid fraction:

$$\text{NCRIF} = \frac{\rho_{\text{s}}(T)}{\rho} = \frac{I(T_0) - I(T)}{I(T_0) - I_{\text{empty}}}$$
(2)

where I_{empty} is the inertia momentum of the empty cell. For bulk liquid helium, $\rho_s(T)/\rho$ tends to unity at low temperature.

2.2. The experiment by Kim and Chan

Kim and Chan studied a TO whose box was an annular channel filled with solid helium [10]. Figure 2(A) shows typical measurements with a pressure P = 51 bar in the helium cell. Below $T_0 \simeq 200$ mK, the resonance period $\tau(T)$ starts to depart from the curve expected by extrapolation of higher temperature data: this was interpreted as the signature of supersolidity. As the change is small, equation (2) can be rewritten as

$$\text{NCRIF} = \frac{\tau(T_0) - \tau(T)}{\tau(T_0) - \tau_{\text{empty}}}$$
(3)



Figure 3. The non-classical rotational inertia fraction (NCRIF) measured by Kim and Chan [10] as a function of temperature for different maximum oscillation velocities (see the legend). This is in a sample ending at 26 bar after growth at constant volume.

assuming a constant G. At low temperature, the NCRIF reaches 1.3%, as if this amount of the helium mass decoupled from the oscillating walls. Note that the high temperature data for $\tau(T)$ extrapolate to a value about 0.2% lower than at T_0 , so that, in equation (3), the extrapolated value of $\tau(T)$ should be used instead of $\tau(T_0)$ when the maximum NCRIF is small. In addition, figure 2(A) shows that the NCRIF depends on the maximum velocity v_{max} during the oscillation $(v_{\text{max}}$ is computed at the mean radius of the channel and in the low temperature limit): at low v_{max} , the curves are identical, but when v_{max} becomes larger than approximately 10 μ m s⁻¹ the NCRIF decreases and finally tends to vanish. This is more clearly seen when the NCRIF calculated from equation (3) is plotted versus temperature, as shown in figure 3. Finally, figure 2(A) shows a broad minimum in the oscillation amplitude: since, for all curves, the cell was excited with a constant ac voltage, this indicates the existence of a maximum in dissipation. This was also seen in TO studies of thin helium films on a planar surface [16-18]. These observations have been presented as further evidence that a fraction $\rho_s(T)/\rho$ of solid helium 4, equal to the NCRIF, becomes superfluid at low temperature, with a small critical velocity of order 10 μ m s⁻¹.

In a first control experiment with high-purity (99.999%) solid helium 3, which is a fermion, no NCRIF was found by Kim and Chan. A second control experiment involved helium 4 in a cell where a barrier was placed across the annular channel in order to block circular flow. Even in a blocked channel, a superfluid should exhibit non-zero NCRIF because some irrotational flow can take place, but it should be strongly reduced if the thickness of the annulus is very small compared to its diameter [19]. Unfortunately, the dimensions of Kim's blocked cell were different from those of the original, unblocked, one: the width was 15 mm instead of 10 and the thickness was 1.1 mm instead of 0.63 mm (in fact, the actual value is yet different²). An extrapolation led Kim and Chan to estimate that an unblocked cell with the same size as the blocked one should show a period shift of 95 ns. Since they

² In a more recent article [20], Clark and Chan explained that the thickness of the original cell used by Kim and Chan was 0.95 mm, not 0.63 mm.

observed 1.4 ns only (figure 2(B)), they considered that the effect of the barrier was to reduce the period shift to 1.5% of its value for the unblocked cell. A calculation with the experimental geometry of the blocked cell led Mueller [21] to 0.8%, a value which was estimated by Kim and Chan [10] as the 'same order' as their experimental one. In the end, it seems to us that the measured value (1.5% if the extrapolation is correct) is significantly larger than the calculated value (0.8%) for the reduction of the NCRIF due to the barrier. Since this experiment is a crucial test of macroscopic quantum coherence in the sample, we hope that this experiment is repeated carefully with blocked and unblocked cells having the same dimensions, and also with more than one thickness.

In their very first series of experiments [11], Kim and Chan studied the NCRIF of solid helium in a cell filled with a porous glass (Vycor). They observed the same features as described above, as if mass could flow in the glass pores despite their very small diameter (typically 7 nm [22]). This is also observed when a Vycor sample filled with *liquid* helium 4 is cooled through its superfluid transition temperature [23], although the critical velocity is much larger than for the solid. In the first supersolid experiment with Vycor, adding helium 3 was found to decrease the NCRIF (it vanishes for helium 3 concentrations above 0.1%), to broaden the transition and to increase its onset temperature T_0 .

Subsequent experiments have confirmed these observations. Solid helium confined in porous gold with characteristic pore diameter 490 nm exhibits a similar behaviour [24]. For bulk solid helium, the pressure variation of the zero temperature NCRIF was later measured with less scatter than in the first experiment, and a maximum was found around 55 bar [25] (see figure 4). Eventually, the effect of adding helium 3 impurities was confirmed in bulk solid helium 4 samples. Starting from ultra-pure helium 4 with less than 2 ppb ³He, Kim *et al* found that T_0 increases monotonically with helium 3 concentration and that the supersolid fraction has a broad maximum around 0.2 ppm [26, 27].

3. Superfluid vacancies?

We now turn to the early theories of supersolidity and to their analysis in more recent theoretical works.

3.1. Early theories

Given these observations, Kim and Chan proposed that solid helium 4 is 'supersolid'. Such a possibility had been considered by Reatto [3], Andreev and Lifshitz [4], Chester [5], Leggett [6], Imry and Schwartz [7] and a few other theorists about 35 years earlier. The argument by Andreev and Lifshitz was simple. Suppose that one creates a classical vacancy by removing one atom on one site of a crystal lattice: there is a cost for this which is the activation energy E_0 of this classical—i.e. localized—vacancy. Suppose now that, by quantum tunnelling, this vacancy can exchange its position with a neighbouring atom. If the exchange frequency ν is large enough, the vacancy becomes a quantum wave which is delocalized in the whole lattice. Its energy lies inside a



Figure 4. The pressure variation of the NCRIF at low temperature, as measured by Kim and Chan in 2004 (open squares [10]) and in 2006 (filled circles [25]). The solid line is a guide to the eye. Kim and Chan labelled the left scale ρ_{S0}/ρ because, in their model, the NCRIF is the ratio of the superfluid density in the T = 0 limit to the total density ρ . In the 2006 configuration, improved growth conditions reduce the scatter in the data and a non-monotonic pressure variation shows up: the supersolid fraction increases up to $P \simeq 55$ bar before decreasing at higher pressure.

certain band whose width is proportional to v. If v is large enough, the bottom of the band has a negative value, so that the crystal is invaded by a finite density of vacancies which are called 'zero point vacancies' because they should exist even at T = 0. Moreover, in a crystal made of Bose particles such as helium 4 atoms, these quantum vacancies are also Bose particles; they should undergo a Bose-Einstein condensation (BEC) at a certain critical temperature T_c and form a superfluid inside the crystal. Since a vacancy moving in one direction is equivalent to an atom moving in the opposite direction, the superfluidity of zero point vacancies would allow mass transport through the lattice without dissipation. The final situation would be the paradoxical coexistence of order in real space (the crystalline order) and in momentum space (the Bose condensate of vacancies). Andreev and Lifshitz have considered the possible propagation of hydrodynamic modes in solid helium 4 if supersolidity were to occur and their work was later extended by Saslow and by Liu [28, 29].

This possibility looked contradictory to the work of Penrose and Onsager, who had introduced the formalism of the density matrix to generalize BEC to interacting systems like liquid helium. The existence of a condensate appeared as 'off-diagonal long range order' (ODLRO) in the one particle density matrix. The physical meaning of ODLRO is that there is a non-zero overlap between two wavefunctions describing the whole system and differing by the exchange of two atoms a distance r apart, even in the limit where r tends to infinity. It implies phase coherence at macroscopic scale. In their 1956 article [30], Penrose and Onsager wrote a paragraph claiming that ODLRO could not exist in crystals. As we shall see, this fundamental issue is rather delicate.

It was noticed by Chester [5] that Penrose and Onsager had used non-symmetrized wavefunctions to demonstrate their result, so that their conclusion was questionable. A few years later, Imry and Schwartz [7] explained that, in a crystal with one atom per site, the absence of ODLRO is not a question of symmetrization but a question of wavefunction overlap. By generalizing arguments by Matsuda and Tsuneto [31], Imry and Schwartz showed that a large class of wavefunctions describe quantum solids which do not exhibit ODLRO in the absence of vacancies [7]. They also showed that some lattice gas models show ODLRO in the absence of vacancies but they are not solids in the usual sense because they allow double occupation of sites. Imry and Schwartz finally showed that, in the presence of vacancies, BEC could occur. Leggett [6] predicted that, in the case of ODLRO, the crystal should exhibit NCRI properties; this is a consequence of phase coherence in the crystal. All known three-dimensional superfluid systems do exhibit ODLRO and NCRI³. In view of the above arguments, there remain at least two fundamental questions to be answered.

- (1) Is it a *universal* result that, without vacancies, crystals are not superfluid?
- (2) Are there vacancies in the ground state of solid helium, in which case could their existence explain experimental observations? In particular, is the 1% NCRIF observed by Kim and Chan the consequence of a finite density of vacancies in the ground state of helium crystals?

This double issue has been reviewed by Prokof'ev [12], but it might be useful to come back to it as briefly as possible. Prokof'ev et al [32-34, 12] define 'commensurate' crystals as crystals where the number of atoms N is equal to the number of lattice sites $N_{\rm s}$. In contrast, crystals are called 'incommensurate' if $N \neq N_s$. According to Prokof'ev and Svistunov [32, 12], commensurate crystals are 'nearly always' 'insulating', that is non-superfluid. The superfluidity of a crystal requires the presence of vacancies or interstitials which do not form bound pairs together. 'Nearly always' means the following: assume that, at a particular density or pressure, a crystal is superfluid and commensurate because the number $N_{\rm vac}$ of free vacancies equals the number of free interstitials N_{int} ; then, since there is no reason for the energy of vacancies to keep equal to that of interstitials under different density or pressure conditions, the crystal would be incommensurate at neighbouring densities. In other words, the probability that a commensurate crystal is superfluid should be negligible. As for the possibility that the ground state of helium crystals is incommensurate, they claim that it is contradictory to various experimental measurements of the activation energy for vacancies or interstitials, also to their own calculations. They mainly consider vacancies because the energy of interstitials is generally found to be larger than that of vacancies (their calculations [34] at the melting pressure $P_{\rm m} = 25$ bar give $E_{\rm vac} = 13$ K and $E_{\rm int} = 23$ K). They further argue that the vacancy-vacancy interaction is attractive, so that a finite density of vacancies at low temperature should lead to their aggregation, that is to a liquid-solid phase separation.

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In a purely classical Monte Carlo simulation, Ma *et al* also find that the vacancy–vacancy and the interstitial–interstitial interactions are attractive so that they should aggregate and phase separate [35]. However, Mahan and Shin [36] calculated the interaction between two *fixed* vacancies within elasticity theory. They found that it is indeed attractive in the basal plane as found in the simulations by Boninsegni *et al* [34] but that it is repulsive in other directions. Mahan and Shin further explain that their calculation should be improved by taking the motion of vacancies into account.

Anyhow, in 1969, Reatto [3] and Chester [5] had shown that, if one describes a crystal with 'Jastrow wavefunctions', BEC should occur. More recently, Galli, Rossi and Reatto [37] have proposed that the ground state of a helium crystal is commensurate *and* superfluid. Their conclusion is based on a variational calculation using 'shadow wavefunctions' (SWFs). They claim that quantum fluctuations create vacancy–interstitial pairs, which provide an exchange mechanism for atoms.

As for Clark and Ceperley, they explain in the long introduction of their article [38] that 'it is not possible to state a general theorem covering whether all quantum crystals must or cannot have BEC'. They argue that, even when variational calculations describe static properties of helium crystals with great accuracy, which is the case for SWF, they may not describe BEC, which is very sensitive to the range of the interatomic correlations. This is why they use path integral Monte Carlo (PIMC) simulations which are based on the Aziz potential and, 'in principle', exact. The Aziz potential is also known to accurately describe the He-He interaction [39]. According to Clark and Ceperley, variational calculations find BEC as long as the interatomic correlations are short ranged as in Galli's work. Their MC calculation shows no ODLRO in commensurate crystals, while an MC calculation using SWF, 'being built from short range correlations, leads inevitably to superfluidity'. Clark and Ceperley also criticize the use of Jastrow wavefunctions because they do not correctly reproduce the rms vibration of atoms around their lattice sites: the Lindemann ratio $\gamma = \sqrt{\langle u^2 \rangle}/a$ (where *u* is the displacement and a is the lattice parameter) has been measured [40] to be 0.26 at the melting pressure. This is much larger than in classical systems, where $\gamma \simeq 0.14$. Cazorla and Boronat [41] use another type of MC calculation, where atoms interact with each other through the Aziz potential, and with a periodic external potential of adjustable height. In the absence of vacancies, they find a non-zero superfluid fraction in a range of potential heights which corresponds to a range of Lindemann ratios that includes the experimental value; the superfluid fraction then amounts to 8%. However, the Lindemann ratio describes an average while what appears to be important for superfluidity is the overlap of wavefunctions, which has to do with the tails of the density profile of the atoms around their site positions. In summary, in our opinion, our first question is still a matter of controversy: there exist model systems which are commensurate and show ODLRO, but it is not yet absolutely clear if these models describe real crystals or only artificialperhaps exceptional-ones.

Let us now come to our second question. As already mentioned above, Boninsegni *et al* [34] calculated activation

³ In two dimensions, there can be superfluidity without ODLRO in the usual sense since the off-diagonal terms in the density matrix decrease algebraically to zero at long distance. As explained by Prokof'ev [12], if one takes an integral definition of ODLRO, one recovers the equivalence with a non-zero ODLRO in superfluid systems, even in two dimensions.

energies with a path integral Monte Carlo method. For vacancies they found $E_{vac} = 13$ K, and for interstitials $E_{int} =$ 23 K (both at the melting pressure $P_{\rm m}$). With a similar method, Ceperley and Bernu [42] had found $E_{int} = 50$ K for interstitials but, according to a note in [34], they later acknowledged that their value may need to be revised. With their variational method using SWFs, Pederiva, Chauduri, Fantoni, Chester and Reatto had found $E_{\rm vac} = 15$ K and $E_{\rm int} = 30$ K at $P_{\rm m}=25$ bar [43, 44]. This result is similar in magnitude to Boninsegni's, and also to an experimental determination by Fraass et al [45], so that one could consider as well established that vacancies have a large activation energy. As a consequence, not only are there no zero point vacancies, but at temperatures of a few hundred millikelvin the density of thermally activated vacancies is negligible. For interstitials, it would be even more strongly established. However, in 2006, Galli and Reatto [46] criticized the results of their own group and claimed that, due to finite size effects, 'the question whether the ground state of bulk solid helium 4 is commensurate or incommensurate is still undecided', but they 'noticed that the ground state is incommensurate for the best variational wavefunction'. To this objection, Boninsegni et al [34] replied that their calculation of either E_{vac} or E_{int} did not show any dependence on the system size. As we shall see now, the theoretical controversy on the value of E_{vac} has progressed thanks to arguments put forward by Anderson, Brinkman and Huse [47], to counter-arguments to the latter, and to new experimental measurements.

3.2. Search for zero point vacancies

In 2005, Anderson, Brinkman and Huse [47] proposed that, in fact, helium crystals contained zero point vacancies. Their argument was based on experimental observations and twofold. They first considered the temperature variation of the lattice spacing $\Delta a/a(T)$ which had been measured by Fraass *et al* [45]. If vacancies are thermally activated, both their density and $\Delta a/a$ should vary as $\exp(-E_{\rm vac}/k_{\rm B}T)$. With this assumption, Fraass *et al* had concluded that $E_{vac} = 8$ K at the melting pressure $P_{\rm m}$, and that it increased with pressure [45]. This is the same order of magnitude as the recent calculations mentioned above [43, 44, 34]. But Anderson et al argued that, in the presence of zero point vacancies, that is if $E_{\text{vac}} =$ 0, $\Delta a/a$ should vary as T^4 . They further showed that the measurements by Fraass et al, which were limited to the region above 0.8 K, could be fitted equally well with an exponential and with a T^4 law, so that, according to them, there could exist some density of zero point vacancies. However, in 2007, Blackburn *et al* [48] measured $\Delta a/a(T)$ in the range 0.14–0.8 K. They found it constant within 10^{-4} . Although unfortunately their measurements could not be compared with those by Fraass et al because their respective temperature domains did not overlap, Blackburn's results are compatible with thermally activated vacancies but not with the T^4 law proposed by Anderson et al for zero point vacancies. If possible, it would be interesting to extend these neutron studies to even lower temperatures, since the recent experiments by Clark, Lin et al [49, 50] indicate a transition temperature around 75 mK (see sections 4.5 and 4.6).

The second argument by Anderson et al concerned the temperature variation of the specific heat C(T). They argued that, if vacancies were thermally activated, the specific heat should have an exponential correction to the usual T^3 phonon But, instead, Gardner *et al* [51] had found a T^7 term. correction. Any phonon term of higher order should scale as the ratio (T/Θ_D) to some power, where Θ_D is the Debye temperature. According to Anderson et al, this ratio should be small because Θ_D is larger than 26 K [51]. Furthermore, they predicted a T^7 correction in the presence of zero point vacancies. However, Maris [52] calculated the higher order phonon terms due to the dispersion of phonons. He found that there is a large coefficient in front of the powers of $(T/\Theta_{\rm D})$ and that it is possible to fit the T^7 term with reasonable values of the phonon dispersion relation. In summary, the suggestions by Anderson et al that helium crystals contain zero point vacancies have certainly triggered an interesting debate but it is not proven that they describe the reality of these crystals.

In the end, one has progressively arrived at the conclusion that, if the anomalies observed in TO experiments are related to the superfluidity of some mass inside helium crystals, the mechanism cannot be as simple as originally proposed in 1969-70. In fact, Andreev's mechanism should lead to the existence of dc superflow in single crystals and, despite several attempts [8], this has never been observed. A more recent search for dc superflow was made by Day et al in two successive experiments [53, 54]. They first studied solid helium confined in a porous glass (Vycor with 7 nm diameter channels). As the pressure was increased outside the Vycor, mass flow into the Vycor could only be seen at a temperature high enough for thermally activated diffusion to take place. No indication of superflow was found at low temperature. In a subsequent experiment, they studied flow through an array of 25 μ m channels with the same negative answer [54]. The only experiment where a dc superflow was observed is the later one by Sasaki et al [55]. We will see below that it required the presence of grain boundaries. In fact, several experiments pointed out the importance of disorder at the same time it was invoked by theorists [32, 38, 12, 56].

4. Disorder

4.1. Sample preparation

After the discovery by Kim and Chan of the NCRI in solid helium, several groups tried to reproduce their experiment. These other groups also found NCRI [57–60], but with some scatter between samples, as already noticed by Kim and Chan. This prompted the study of annealing effects. Annealing is performed by warming up the sample close to the melting point for some time (typically 10 h), and cooling down slowly afterwards. Annealing is a well known method to improve the quality of crystals. Rittner and Reppy [57] annealed their samples for 13 h between 1.4 and 1.5 K, the melting temperature at 26 bar. After annealing, the magnitude of the NCRIF decreased below the noise level of their detection (see figure 5). Although Penzev *et al* reported the observation of a 10% *increase* of the NCRI after annealing [58], and



Figure 5. Rittner and Reppy [57] have found that annealing the helium sample reduces the period shift of their torsional oscillator below the noise level in their experiment. The first run (upper panel, upward triangles) shows a 20 ns period shift. Upon partial annealing, the period shift is decreased (lower panel, downward triangles). After the solid is melted and refrozen (upper panel, full circles), a signal similar to that in the first run is recovered. Finally, a full annealing of the sample (lower panel, stars) reduces the NCRIF below the detection level.

Kondo *et al* found no effect of annealing [59], it is now generally accepted that annealing reduces the disorder and consequently the magnitude of the NCRI, especially since the careful analysis of annealing by Rittner and Reppy [61] and by Clark, West and Chan (see section 4.5) [49]. Note that differences between experiments may come from each cell having pinning centres of variable strength. Furthermore, an increase of the NCRI could be the consequence of the plug moving along the fill line during the annealing, a possibility which was not investigated in all experiments.

The link of supersolidity to disorder was further supported by Rittner and Reppy, who studied rapidly cooled samples. When their solid helium sample was quickly grown by quenching the cell down in temperature, they observed a larger NCRIF [57]. In a later series of experiments with thinner sample space in the torsion bob, Rittner and Reppy obtained an NCRIF as large as 20% [62]. By reviewing other TO experiments [25, 58–60], they further noticed that the NCRIF increased monotonically with the surface to volume ratio of the cell (see figure 6). In fact, this ratio is about the inverse thickness of the sample because the empirical law applies to cells which are all cylindrical. Experimental data corresponding to porous glass [11] or to porous gold [24] lie far away from the others on this graph, indicating a different mechanism. Rittner and Reppy proposed that, as the sample thickness decreased, solidification could proceed more quickly and produce more defects (dislocations, grain boundaries...), which could also be more efficiently pinned to walls.

This series of experiments pointed out the importance of disorder resulting from the sample preparation. In TO experiments where the sample space is suspended by a small rod, it has generally a poor thermal contact with the lowest temperature part of the refrigerator, and samples are usually



Figure 6. The variation of the NCRIF, i.e. the ratio of the superfluid density ρ_s in the T = 0 limit to the total density ρ , as a function of the surface to volume ratio, according to Rittner and Reppy [62]. In addition to their own measurements (solid circles and downward filled triangles), they have included the results from Chan's [10] (open circles, open triangle, and open square) and Shirahama's groups [59] (filled upward triangle). Measurements in porous materials (Vycor and porous glass) are not included because they lie far outside this graph.

grown at constant volume. For this, the cell is first filled at high temperature and at some pressure above 50 bar before it is cooled down. Since the cell is not the coldest point, a solid plug forms somewhere along the fill line, presumably near the 1 K pot, which is usually cooled down first, before the ³He-⁴He mixture is circulated in the dilution unit and millikelvin temperatures can be reached. This plug isolates a certain mass in the cell, which is then cooled down at constant volume V (if the plug does not move, of course). When the cell temperature reaches the solidification line $P_{\rm m}(T)$ in the phase diagram, crystallization starts and, as T keeps decreasing, the pressure in the cell also decreases along the melting line $P_m(T)$. As a result, the pressure is not constant during crystallization, so that, although crystallization takes place in a time of order one hour, the resulting solid is usually polycrystalline. This was verified by Sasaki et al, who could observe crystals grown at constant V in their optical cryostat [63] (see figure 7). Sasaki et al did not see any significant difference in the amount of disorder when the solidification proceeded through the bcc-hcp transition. When crystallization was fast from the normal liquid phase, Sasaki et al found dendritic growth, as had been already observed by the groups of Hallock [64] and Okuda [65]. In order to obtain single crystals one needs to grow the solid phase by adding mass at constant pressure and temperature through an open fill line. This can be done easily from the superfluid liquid if the cell is the coldest point along the helium path [66], or from the normal liquid with appropriate temperature gradients.

4.2. Grain boundaries

In 2006, Sasaki *et al* [55] studied dc mass flow through a macroscopic solid sample inside a glass test tube (1 cm inner diameter). The tube was placed upside down in a helium cell, it was vertical and closed at the top. Solid helium was grown from the superfluid at a temperature of 1.3 K, and a sample was



Figure 7. In their optical cryostat, Sasaki *et al* [70, 63] could grow solid helium samples at constant volume in a time of order one hour as usually done in TO experiments. This photograph shows that the sample is polycrystalline. The window is 11 mm wide. The grain boundaries are revealed by lowering the pressure to the melting pressure $P_{\rm m} = 25.3$ bar so that the contacts of grain boundaries with the cell windows are invaded by liquid helium (see section 4.2).

prepared with the liquid–solid interface higher by 1 cm inside the tube than outside. Inside such a cell, where the temperature is highly homogeneous, gravity is relevant so that the solid occupies the bottom of the cell, under the liquid, which is less dense. Liquid–solid interface levels can change by melting in one place and crystallizing in another place. Now, since the solid density is 10% larger than the liquid one, the solid could melt inside the tube only if some mass could escape. Since the top of the tube was closed, mass had to flow through the solid.

Several samples had no apparent disorder. In particular, their interface with the liquid phase showed no grooves, indicating the absence of grain boundaries. For such samples, which were probably single crystals, no relaxation of the interface took place within 50 μ m in 4 h. This result implied that, in these samples, the product $(\rho_s/\rho_c)v_c$ had to be less than $3 \times 10^{-4} \,\mu \text{m s}^{-1}$ (ρ_{s} and ρ_{C} are respectively the supersolid density and the total solid density). This result is 300 times less than expected from the values obtained in the TO experiments $(\rho_{\rm s}/\rho_{\rm C} \simeq 0.01$ and $v_{\rm c} \simeq 10 \ \mu{\rm m\ s^{-1}}$). It shows that there is no measurable density of mobile vacancies in single crystals. Either helium single crystals are not supersolid or the mechanism for supersolidity is not as simple as originally proposed by Andreev and Lifshitz [4]. One should also notice that Sasaki's observations were made at the solid-liquid equilibrium where crystal properties could be different from the ones at higher pressure. For example, it is possible that defects anneal out more efficiently at the melting pressure. This result is also at odds with the hypothesis by Khairallah and Ceperley [67] and Dash and Wetlaufer [56] that a superfluid film exists at the interface between glass walls and solid helium. Of course, the experiment gives only an upper bound for the mass flow, so that this hypothesis is not totally ruled out, but it was also found to be contradictory to their own results by Day and Beamish, who searched for dc mass flow along small capillaries [54] (in their previous experiment [53], they found no dc mass flow through Vycor, but in that particular case one could object that mass flow required a deformation of the crystal lattice, not only a flow though a fixed lattice, so that the results were not as conclusive as in their later experiment).

Sasaki et al found a different behaviour with polycrystalline samples [55]. Fast growth of the solid led to the formation of grain boundaries (GBs), whose presence was revealed by the existence of grooves at the solid-liquid interface. It is well known in materials science [68] that these grooves result from the mechanical equilibrium between the GB surface tension σ_{GB} and the liquid-solid interfacial tension σ_{LS} . After growth, some GBs moved away and some remained, presumably pinned to walls. In three samples with GBs, mass could flow and the interface relaxed [55] inside the glass tube. This relaxation was not exponential as it would be for a classical system. On the contrary, it took place at a constant velocity, as expected for a superflow at its critical velocity, which is independent of the applied force (here a height difference). At first sight, Sasaki's results looked like evidence for the existence of superflow along GBs connecting the liquid inside the tube to the one outside the tube. If true, Sasaki's measurement with only one apparent GB could be understood with a critical velocity along the GB of order 1 m s⁻¹. This value is similar to what had been previously found for liquid films of atomic thickness [69]. Note that, in the experiments by Day et al [53, 54], the absence of superflow could be due to the absence of connection between GBs along very thin tubes. The same absence of connection probably explains why Sasaki et al found that mass did not flow along the capillary (0.6 mm in diameter) connecting their pressure gauge to their experimental cell [70].

GBs have been predicted to be superfluid by Burovski *et al* in the frame of a general model [71]. Pollet *et al* made a more precise calculation adapted to the particular case of helium [72]. They found that superfluidity should occur in GBs at temperatures of about 0.5 K, except for special relative orientations of the grains (see figure 8). In their experimental study, Sasaki *et al* [55] had found superflow at 1.13 K and it might exist at even larger temperature. They proposed that, perhaps, GBs were thick enough on the melting curve for their superfluid transition temperature to be close to that of bulk liquid helium. However, a later experiment by Sasaki *et al* [73] led to another possible interpretation of their flow experiment, as we shall see now.

Sasaki *et al* studied the boundary between two stable grains, and measured the dihedral angle 2θ of the groove at the top of the GB (see figure 9). The GB energy can be deduced from the relation $\sigma_{GB} = 2\sigma_{LS}\cos\theta$, where σ_{LS} is the known energy of the liquid–solid interface [66]. They found θ around 13° and strictly positive, meaning that the thickness of GBs is not macroscopic (even at the liquid–solid equilibrium, the GBs are not completely wetted by the liquid phase). They also found that the line of contact of GBs with solid walls is in reality a liquid channel [73] (see figure 10). This channel has a triangular cross-section, which is stable if $(\theta + \theta_c) < \pi/2$, where θ_c is the



Figure 8. Pollet *et al* [72] have simulated a GB between two crystals with different orientations and a pyramidal shape, in equilibrium with liquid regions on each side (upper panel). The density of black dots (lower panel) indicates the local superfluid density: it is found to be non-zero in the liquid of course, but also inside the GB.

contact angle of the liquid–solid interface with the wall (see figure 10). The channel width w is inversely proportional to the depth z below the flat part of the liquid–solid interface, as can be understood with a simple capillary model [73]. Typically, $w = 20 \ \mu \text{m}$ at $z = 1 \ \text{cm}$. Sasaki *et al* finally predicted that, except perhaps in the presence of inhomogeneous stresses, these channels should be of atomic size at about 35 bar ($P_{\text{m}} + 10 \text{ bar}$), and vanish above this pressure.

This suggests another interpretation of the test tube experiment: mass could flow along the liquid channels where GBs meet the glass walls, not necessarily along the GBs themselves. If true, it implies a critical velocity of order 3 mm s⁻¹ along the channels, a value which is comparable to what has been found in capillaries [74]. In order to decide what is the right interpretation and if the inside of the GBs is really superfluid or not, the ENS group needs to build a new version of the tube experiment. Note that, since $2\theta < 60^\circ$, there should



Figure 9. A GB makes a groove when emerging at the liquid–solid interface. From the groove dihedral angle 2θ and the value of the liquid–solid interfacial energy σ_{LS} , Sasaki *et al* [73] obtained the GB energy $\sigma_{GB} = 2\sigma_{LS} \cos \theta$. This photograph also shows that the contact lines between the GB and each glass window of the cell are liquid channels whose width is a few tens of micrometres, depending on the depth below the liquid–solid interface on top.

be similar liquid channels inside polycrystalline samples at all GB crossings [63].

4.3. Glassy or liquid regions

As mentioned above, rapidly quenched samples show large NCRIF [57]. In a particularly thin sample cell, it has been found that helium solidifies very fast and the NCRIF reaches 20% [62]. After annealing such samples with high NCRIF, Rittner and Reppy found that the pressure in the cell decreased by several bars, something which had also been noticed in the early experiments by Fraass et al [45]. This is a strong indication that quenched samples are inhomogeneous in density. They must contain regions with a low density which can crystallize during the later annealing so that the pressure, which is measured on one side of the cell, relaxes down. In reality, this 'pressure' is the normal component of a local stress tensor, which does not have to be isotropic or homogeneous in space. Of course, this interpretation supposes again that, during the annealing, the plug which blocks the fill line of the cell does not move. The inhomogeneity in density is supported by the measurements of Grigor'ev et al [75].

They cooled down solid helium samples which had first been quickly grown at constant volume. They measured the temperature variation of the sample pressure, in reality a local stress component again. Grigor'ev *et al* measured what can be called a solid isochore $P_V(T)$. As in Rittner's experiment, they observed a decrease in pressure after annealing as if part of the sample had recrystallized. Furthermore, they found that $P_V(T)$ varied as $AT^4 + BT^2$. The T^4 term was expected, since there must be a contribution from phonons. The T^2 term could be consistent with glassy regions whose entropy should be linear in T, and it disappeared after annealing. Liquid regions



Figure 10. (a) Three-dimensional view of the contact between a grain boundary (dash–dotted line) and a wall. The hatched area shows the contact of the wall with the solid. (b) Horizontal cross-section of the liquid channel near the wall.

could also exist if the stress is highly inhomogeneous, not only because the effective pressure could be low in particular regions of the sample. This is because, in a solid, nonhydrostatic stresses add elastic terms in the energy, so that the equilibrium with the liquid phase is displaced towards higher pressure. However, in order to lead to a T^2 term in the isochore, these liquid regions would need to be one dimensional. Indeed, consider a *d*-dimensional object whose density is δ_d , meaning that in a volume *V* there is a length, surface, or volume $\delta_d V$ of such objects, for d = 1, 2 or 3, respectively. The phonon gas in this object gives a contribution to the pressure which can be estimated from $F_{\rm ph}$, the phonon contribution to the free energy:

$$P(T) - P(T = 0) = -\left(\frac{\partial F_{\rm ph}}{\partial V}\right)_{\rm T} = -\frac{F_{\rm ph}}{V}$$
$$= \delta_d \, k_{\rm B} T \int_0^{\omega_{\rm c}} \mathrm{d}\omega \, n(\omega) \ln\left[1 - \exp\left(-\frac{\hbar\omega}{k_{\rm B} T}\right)\right] \tag{4}$$

where ω_c is a cut-off frequency corresponding to a wavelength of the order of the interatomic distance, and $n(\omega)$ is the density of states of the *d*-dimensional phonon gas. At low temperature $(T \ll \hbar\omega_c/k_B)$, this leads to $P(T) - P(T = 0) \propto T^{d+1}$.

Grigor'ev et al claimed that the magnitude of their T^2 term was consistent with 5% of their sample being glassy solid helium. Could this explain the TO results? It is possible that, in Rittner's samples, which were quenched even faster than those of Grigor'ev, there is more than 5% glassy regions and this would need to be checked. Boninsegni et al have simulated a helium glass and found that it should have a transition to a superfluid state which they called a 'superglass' [33] (see figure 11). One should also notice that a grain boundary with a large orientation difference between the two adjacent grains can be viewed as a two-dimensional helium glass. According to Pollet et al it should be about three atomic layers thick, a result which is consistent with Sasaki's measurements since GB grooves have a non-zero dihedral angle. One interesting question is whether the glassy regions in Rittner's experiment would need to be connected in order to provide a macroscopic phase coherence in the whole sample. They could be in



Figure 11. Boninsegni *et al* [33] have calculated the pair correlation function g(r) in hcp helium crystals and in a glass with the same density (upper panel); they have also calculated the off-diagonal term n(r) in the superfluid state of the glass compared to the hcp crystal at the same temperature and density (lower panel). The superglass state exhibits ODLRO: the off-diagonal term tends to a non-zero constant at large distance, while in the crystalline state it tends to zero exponentially.

contact with each other or be connected by grain boundaries. Many theories rely on the hypothesis that there is indeed a macroscopic phase coherence in the sample. As mentioned in section 2, the experimental check of phase coherence by Kim and Chan needs to be repeated, especially in thin samples, where the reduction in the NCRIF should be drastic. Andreev further discussed the properties of supersolid glasses in terms of two-level systems [76]. Whether these two works are relevant to explain Rittner's observations of very large NCRIF is, in our opinion, an open question. It is also possible that, for experiments in Vycor or in porous glass, there is a large contribution from the disordered region at the interface between walls and solid helium and, according to Dash and Wetlaufer [56], and Khairallah and Ceperley [67], this disordered region is superfluid at low T.

4.4. ³He impurities

Already in their first article [11] Kim and Chan noticed that the presence of ³He impurities had a strong effect on the anomalies observed in their TO experiments, even at concentrations of order 10^{-6} or less. They checked this further [26, 27] and found that T_0 increases monotonically with ³He concentration, while the NCRIF had a broad maximum around 0.2 ppm. This high sensitivity was soon considered by Beamish [77] as a strong indication that, whatever they are, the TO anomalies had to do with defects such as dislocations and GBs, where ³He impurities were known to bind. On dislocations, Iwasa had measured a binding energy of 0.6 K [78, 79] and Paalanen had found 0.7 K [80]. At a temperature which should depend both on ³He concentration and on dislocation line density, the impurities should attach to dislocations where their local density should become much larger than the average one in the bulk. Once decorated with ³He atoms, the dislocations should behave differently: their possible superfluidity (see section 4.5) should change, as well as their mobility, consequently their Since GBs can be considered as sets of fluctuations. dislocations, ³He impurities should also bind to them and change their properties. It is also possible that ³He impurities bind to the crossing nodes or lines between dislocations or GBs and consequently affect their connections. As we shall see below, more measurements have been performed in the respective groups of Beamish [81] and Chan [50]. They show a sensitivity to ³He concentration down to the 1 ppb level.

4.5. Dislocations in single crystals

Given all the uncertainties in the structure of samples grown at constant volume, Clark, West and Chan have made a new series of measurements in samples grown at constant T and P from the superfluid [49]. It is well known [66] that, when grown slowly from the superfluid, by adding mass in the cell which stays at the liquid–solid equilibrium pressure $P_{\rm m}(T)$, solid helium samples are usually single crystals.

By using ultra-pure samples (1 ppb ³He only), Clark *et al* observed NCRIF in the range 0.03–0.3% with an onset temperature $T_0 = 75$ mK, lower than with samples of natural isotopic purity (0.3 ppm), where $T_0 \simeq 150$ mK. According to Clark *et al*, the transition in their samples is sharper and agrees with a power law of the form $A(1 - T/T_c)^{0.67}$, as expected for a superfluid transition (with T_c a true critical temperature equal to 60 mK, while the onset temperature T_0 would be related to finite frequency effects). Clark *et al* further noticed that for crystals grown at constant volume a succession of annealing cycles progressively moved the transition curve

down to the one corresponding to single crystals grown at constant pressure.

Clark's experiment first showed that, at least in such samples, the NCRI could not be explained by the superfluidity of GBs because their samples contained either no GBs at all or very few. If one supposed that the superfluid fraction is due to superflow along grain boundaries, the typical grain size would need to be rather small: if, for simplicity, one considers grains forming a cubic lattice of period *l*, the GB density is 3/l; if each GB has an effective superfluid thickness α , the grain size *l* corresponding to a superfluid fraction ρ_s/ρ is

$$l = \frac{3\alpha\rho}{\rho_{\rm s}}.$$
 (5)

For $\rho_s/\rho = 0.1\%$, one finds a typical grain size $l = 0.1-1 \ \mu m$ if α is 0.1–1 interatomic distance a = 0.3 nm. This is perhaps possible with polycrystalline samples grown from the normal liquid, where Sasaki *et al* found grain sizes smaller than 10 μ m (see figure 7), but it is obviously not possible with crystals grown slowly from the superfluid [66].

Clark et al compared their results to various models. They considered the 'vortex liquid' proposed by Anderson [82]. As also proposed by Day and Beamish [81] (see section 5.2 below), helium 3 impurities could condense on dislocation networks and pin both vortices and dislocations. These ideas are further developed in the recent article by Kim et al [27]. The interplay of vortices, dislocations and impurities would need to be included in a more complex model. In the crystals studied by Clark et al, the dislocation density could vary by several orders of magnitude from one sample to another because of sensitivity to growth conditions. In addition to growth conditions, it is possible that, after growth, when the samples are cooled down at constant volume and no longer at the liquid-solid equilibrium, non-hydrostatic stresses appear and create more defects. Clark et al found that their NCRIF was 0.3% in one cell but 0.03% in another cell although the two cells were similar: about the same size and the same resonance frequency, the main difference was in the rod material, which was BeCu in one case and AgCu in the other case. The large difference in NCRIF between the two cells shows once more the importance of uncontrolled disorder in experiments, even when crystals are grown with extreme care.

Clark *et al* [49] also compared their results with a scenario based on the superfluidity of dislocation cores. This superfluidity had been considered by Shevchenko [84] and further developed by Toner [85]. The Monte Carlo calculations by Boninsegni *et al* [83] show that the core of *screw* dislocations (not *edge* dislocations) shows phase coherence (see figure 12). As known for a one-dimensional system, this coherence extends over a distance ξ which is inversely proportional to temperature. As a consequence, if there is a dislocation network with a typical distance *L* between dislocations, and if $\xi > L$, the phase coherence extends over the whole crystal.

Boninsegni *et al* argue that, for a given dislocation density $n_d \propto 1/L$, the superfluid mass density ρ_s inside dislocations should scale like T_c^2 . The inside of screw dislocations is found to be a true 1D supersolid: matter is periodically



Figure 12. Boninsegni *et al* [83] have shown that the core of screw dislocations is superfluid. The left picture shows a transverse section of such a screw dislocation, where the density of dots is proportional to the superfluid fraction. The right picture shows a longitudinal section. In both pictures, one sees that the density inside the core follows the crystal periodicity.

distributed in space but also shows long range phase coherence. However, Boninsegni's model is not sufficient to explain Clark's observations for two reasons.

First, Clark *et al* have found that the NCRIF does not vary proportionally to the square of the onset temperature T_0 . Second, a very high dislocation density would be necessary to explain the magnitude of the NCRIF. Even a 0.03% supersolid fraction would need a very high n_d if the only mechanism was flow of mass along the dislocation cores. The argument is similar to the one used to derive equation (5): if dislocations form a cubic network of period l, and if each dislocation contains the equivalent of one dense line of atoms with a diameter a = 0.3 nm, the dislocation density is

$$n_{\rm d} = \frac{3}{l^2} \approx \frac{\rho_{\rm s}}{\rho a^2} \tag{6}$$

which gives 3×10^{11} cm⁻²(1 = 100*a*) for a supersolid fraction of 0.03%. Such a density is perhaps not impossible but it looks very high.

We finally wish to consider another model based on dislocation properties, which has been proposed by Biroli and Bouchaud [86]. They consider the transverse fluctuations of dislocations, which had been ignored by Boninsegni et al, and they propose that, below a temperature T_k , the free energy of kink-antikink pairs on dislocations should become negative. As a consequence, kinks and antikinks should proliferate so that dislocations should become highly mobile and allow atom exchange, which leads to the supersolidity of the whole crystal. According to them, a critical temperature T_c of 100 mK would be consistent with a much lower dislocation density than if transverse fluctuations of dislocations had been neglected. In this model, the increase in line density of dislocations as T decreases should switch on a linear term in the specific heat [87], which needs to be calculated and does not seem consistent with the measurements by Lin *et al* [50] (see below).

4.6. Thermodynamic properties

Assuming that the NCRIF observed in TO experiments corresponds to a real phase transition, it should also be seen on thermodynamic properties. Dorsey *et al* [88] have predicted that, if the normal solid to supersolid transition is of second order, an anomaly in the specific heat should be observed, similar to the well known λ -anomaly of superfluid helium 4. To check this issue, Lin, Clark and Chan looked for a peak in the temperature variation of the heat capacity of solid helium [50].

They used an ac-calorimetry method at 0.1 Hz to study solid samples grown at constant volume. In order to improve the crystal quality, the growth took place over a very long time (20 h) so that their samples were insensitive to any further annealing. From the measured total heat capacity, they subtracted the empty cell contribution (reduced to only onetenth of the total thanks to a cell made of pure silicon) and the T^3 contribution from the phonons in solid helium. Eventually, the possible contribution from ³He impurities was carefully analysed in samples with higher impurity concentrations and found to be negligible for a 1 ppb concentration. The whole procedure led to a peak in the specific heat, centred around 75 mK (see figure 13). This temperature is the same as the onset temperature found by Clark et al [49] with ultra-pure, single crystalline samples. The amplitude of the peak is about 2.0 μ J mol⁻¹ K⁻¹. Given this number and assuming that the transition is a 3D superfluid transition, the expected superfluid fraction can be calculated with the Josephson relation [89] and the two scale factor universality class hypothesis [90, 91]. It scales as $A^{1/3}T_c$, where A is the amplitude of the singular part of the heat capacity per unit volume [92]. Taking for this quantity the peak height and comparing to liquid helium 4, Lin et al estimate the supersolid fraction to be about 0.06%, in agreement with previous NCRIF measurements. Together with its interpretation, this measurement strongly supports



Figure 13. Heat capacity measurements by Lin *et al* [50] of solid helium 4 samples grown at constant volume and containing different helium 3 concentrations (see legend). The temperature variation shows a maximum around 75 mK when contributions from phonons and from helium 3 impurities are subtracted. The dashed lines indicate the standard deviation for the 1 ppb data.

the existence of a true phase transition associated with the anomalies observed in TO experiments.

In their analysis of preliminary heat capacity data [93], Balatsky et al [94] calculated the entropy near T_c , and argued that it was an order of magnitude too small when compared to an estimate based on the entropy $S_{\lambda} = 4.6 \text{ J K}^{-1} \text{ mol}^{-1}$ of helium 4 at $T_{\lambda} = 1.8$ K and high pressure [95]. They used a scaling argument, taking the entropy at the transition to be proportional to the transition temperature, and a 1% supersolid fraction. If we follow their reasoning with the more recent values from [50], we would expect $S(T_c) =$ $4.6 \times 0.075/1.8 \times 10^{-4} = 115 \ \mu J \ K^{-1} \ mol^{-1}$, still larger than the measured 20 μ J K⁻¹ mol⁻¹. However, this scaling argument is questionable: for superfluid helium 4, S_{λ}/T_{λ} is not constant: it decreases by 11% between the saturated vapour pressure (where $T_c = 2.17 \text{ K}$) and the freezing pressure (where $T_{\rm c} = 1.76$ K) [95]. Furthermore, Balatsky *et al* [94] proposed that, in the specific heat, the presence of a term linear in T is the signature of a glassy component in the solid. The calorimetry experiment of Lin et al [50] rules out such a term, but it could exist in more disordered samples. In fact, a linear term that decreases after annealing was reported by Franck in 1964 [96] and interpreted as the contribution due to the vibrations of a dislocation network [97]. It may be that Franck's samples had a much larger dislocation density than Lin's.

Lin *et al* compared their results with the measurement of the melting curve $P_m(T)$ by Todoshchenko *et al* [98]. Indeed, a superfluid transition should affect the solid entropy and consequently the shape of the melting curve. However, after elimination of an artefact due to changes in the elastic properties of the flexible part of their pressure gauge [99], Todoshchenko *et al* found no deviation from the usual T^4 law, which one expects if both the liquid and the solid entropy are dominated by phonons. One possible explanation could be that, being grown at low temperature from the superfluid liquid and being kept on the melting curve, Todoshchenko's samples are single crystals with a very low density of dislocations [66]. In contrast, Lin's samples, being grown at constant volume from the normal liquid, are likely to be polycrystals with a large density of defects (GBs and dislocations), even if they show no change under annealing: if strongly anchored on walls, defects do not anneal out. If true, this would be consistent with the hypothesis that, whatever the exact mechanism, supersolidity requires the presence of defects.

5. Mechanical effects

5.1. The experiment by Day and Beamish

Since a solid differs from a liquid by the non-zero value of its elastic shear modulus μ , it appeared important to measure this quantity in solid helium and to look for a change around 100 mK. This is what Day and Beamish have done with two different methods [81]. They used a pair of transverse piezoelectric transducers, parallel to each other, separated by a thin gap (180 μ m) filled with solid helium. In a first experiment, a quasi-static (20 Hz) voltage is applied to one transducer and produces a shear strain in the neighbouring helium. The other transducer measures the stress associated with this strain, so that μ can be deduced. The same transducers could be used in a second experiment to excite an acoustic resonance near 8000 Hz in the whole helium Usually, an acoustic wave is not purely transverse cell. because acoustic modes can be purely transverse or purely longitudinal only in high symmetry crystalline directions. But any variation of the shear modulus should affect the frequency of the resonant mode. With both methods, Day and Beamish have measured a change in μ which is highly reminiscent of the anomalies observed with TOs (see figure 14).

As T decreases below about 100 mK, μ is found to increase. The effect is large, typically 10%. It starts around 200 mK (100 mK for ultra-pure samples) and saturates below about 50 mK (20 mK for ultra-pure samples; see figure 14). Associated with this change in μ , there is a peak in the attenuation of sound, which is similar to that of the TO. The experiments used strains down to 2.2×10^{-9} , that is one order of magnitude smaller than the plasticity threshold found around 2×10^{-8} . Day and Beamish have also observed annealing and hysteresis effects which are reminiscent of the observations by Aoki et al [60] (see section 6 below). Eventually, the change in μ was found to be sensitive to small ³He concentrations in a way similar to the NCRI in TO experiments: the addition of small amounts of ³He increases the onset temperature below which μ varies and decreases the amplitude of the variation. Given this series of similarities between TO and μ measurements, it is tempting to think that the two types of anomalies have a common physical origin. However, if it exists, this common origin is not yet clear.

5.2. Interpretation and relation to other experiments

Day and Beamish have proposed an interpretation of their measurements in terms of ³He impurities binding to dislocations with a binding energy $E_3 = 0.6$ K. This is the value that had been measured by Iwasa [78, 79] from sound velocity and sound attenuation measurements. Paalanen *et al* [80] had found a similar value $E_3 = 0.7$ K. Note that



Figure 14. The temperature variation of the shear modulus μ as measured by Day and Beamish [81] (filled symbols). For comparison, Day and Beamish have also plotted the NCRIF measurements by Kim and Chan [10] (open circles on the right, 300 ppb) and by Clark *et al* [49] (open squares on the left, 1 ppb). Data have been normalized to the value at 18 mK in order to make the comparison clearer (see [81]). As indicated in ppb, the three sets of data correspond to different concentrations in ³He.

Paalanen et al used a TO method similar to Chan's, except that their oscillator was reduced to a small rod (0.4 mm inner diameter, 1 mm outer diameter) with no cell around, solid helium only inside the rod. Paalanen's TO had a resonance frequency of 331 Hz. He used strain levels larger than 10^{-7} , which are, in principle and according to Day and Beamish, well above the plasticity threshold. He measured changes in μ which were much larger than those of Day and Beamish and it would be interesting to see if this discrepancy is to be attributed to the larger strain amplitude. In order to find $E_3 = 0.7$ K, he used hypotheses similar to those of Day and Beamish, based on the Granato-Lücke theory [87]. In this common model, helium crystals contain a certain density of dislocations which form a network with nodes. The typical distance between two nodes is L. It is also the average distance between dislocations. A dislocation line can resonate like a string of length L at a frequency which depends on L and on the energy per unit length—or line tension—of the dislocation. At resonance, an acoustic wave propagating through the whole system is attenuated. Depending on frequency, the dislocation line moves with the wave or not. Since the mobility of a dislocation depends on temperature, a change in sound velocity should be observed as samples are cooled down, and this is what Iwasa on the one hand and Paalanen on the other hand have indeed measured.

Before considering the interpretation given by Day and Beamish in more detail, we wish to mention the work by Goodkind, who observed changes in the velocity and attenuation of sound in solid helium, which might also be related to changes in dislocation mobility, although Goodkind *et al* interpreted them in terms of BEC *above* a critical temperature of order 200 mK [100, 101]. In another experiment, Mukharsky *et al* used a flexible membrane to excite and detect sound resonances in a solid helium sample [102]. They found resonances around 500 Hz, which shifted down in frequency as *T* was lowered below 100 mK in some samples. These effects were sensitive to annealing and a study as a function of the drive amplitude showed non-linear effects which might be analogous to those observed in TO experiments. These resonances are much too low in frequency to correspond to acoustic modes and their nature needs to be explained. As for acoustic modes, Mukharsky *et al* observed them at much higher frequencies (17–32 kHz), which increased as *T* decreased. This temperature variation is reminiscent of the stiffening observed by Day and Beamish.

Day and Beamish suppose that ³He impurities bind to dislocations below a temperature which depends on both the dislocation density and the ³He binding energy. Suppose further that, once bound, ³He atoms pin the dislocation motion. If the distance between bound ³He atoms is smaller than L, the distance between two nodes which are also supposed to pin the dislocations because they do not slide, then the adsorption of ³He reduces the dislocation motion. The final result should be an increase of the effective μ because moving dislocations provide some strain, which is suppressed if the dislocations are pinned. This model and the corresponding explanation of the measurements by Day and Beamish are very physical and sound very likely, but one should notice that they rely on two hypotheses which would need to be checked, namely that nodes in the dislocation network do not move and that ³He atoms pin the dislocations instead of moving with them. Effects associated with a change in the mobility of dislocations have also been considered by de Gennes [103].

5.3. Comparison with TO experiments

A difficulty appears when comparing with TO measurements. Indeed, one could expect that, when a solid becomes superfluid, it becomes more fluid, meaning that its shear modulus *decreases*, while Day and Beamish have found that it *increases*. Could it be that the change in TO period comes from an increase in the elastic constant G of equation (1), more than from a decrease in the inertia I? Given the very small contribution of the helium elasticity to G, which is dominated by the metal box and rod, this possibility looks surprising but not impossible [20, 81]. Biroli and Bouchaud propose

that a superfluid transition is associated with a proliferation of dislocation lines. They expect a solid sample with more dislocations to be stiffer, just like a polymer tangle with a larger polymer density. They further explain that ³He could have two opposite effects: it could reduce the superfluid density since ³He atoms are Fermi particles which add to the normal component as well documented in the case of superfluid *liquid* helium 4. But it could also increase the number of quenched-in dislocations or even change the energy of kink–antikink pairs on dislocations, which would have the opposite effect.

Let us finally note that Dorsey *et al* [88] had predicted that superfluidity and elasticity should be coupled in a supersolid, so that there should be a singularity in the temperature variation of elastic moduli similar to that of the specific heat. However, what is observed by Day and Beamish is a step, not a peak, in the temperature variation of the shear modulus, and the effect is much larger in amplitude than the broad peak observed by Lin *et al* [50] for the specific heat.

6. Additional remarks and questions

6.1. Analogy with quantum gases in optical lattices

Supersolidity has also been studied with quantum gases of cold atoms in optical lattices (see e.g. [104–108]). There are analogies and fundamental differences between real crystals and such periodic systems. An optical lattice is a structure made with interfering laser beams, in which the light intensity is periodic in space. When a gas of cold atoms is introduced, it interacts with the light and the optical lattice acts as a external periodic potential. The atom-atom interaction is repulsive at short range, that is on each lattice site, with an energy U. There is a kinetic energy J due to tunnelling from site to site, which can be tuned by varying the amplitude of the interfering laser beams. This system is a realization of the 'Hubbard model' [108]. For Bose atoms, it undergoes a transition from a localized state called a 'Mott insulator' when the ratio U/J is large to a delocalized Bose-condensed state when it is small. In other words, the system becomes superfluid if there is enough exchange between atoms.

Greiner et al [104] observed this transition with an average density in the range 1-3 atoms per site. They found a transition around U/J = 36. Their result is consistent with the mean field calculation of a transition at 5.8z, where z is the number of nearest neighbours (z = 6 in a simple cubic lattice) [108]. In reality, the transition is highly sensitive to the filling factor of the lattice n, with singularities at each integer value of n. The commensurate system with n = 1 is always localized for weak exchange (U/J > 5.8z) and always superfluid for strong exchange (U/J < 5.8z). But for infinitesimal deviations from commensurability $(n = 1 \pm \epsilon)$ the system is not fully localized: it contains a small density of 'particles' or 'holes', that is interstitials or vacancies in the language of crystals, whose superfluid transition temperature depends on their density. In the limit where ϵ tends to zero, the superfluid transition temperature goes to zero as well.

Compared to real crystals, quantum gases in optical lattices have several peculiarities. First of all, the filling of sites

with atoms can be adjusted independently of the amplitude of the lattice potential, which is due to the laser beams, not to atom-atom interactions. Second, the trapping needs a wide quadratic potential to be superimposed on the optical lattice, so that the density cannot be homogeneous in the whole system. As a result, what is seen when the transition to a Mott insulator occurs is the appearance of concentric domains with filling factors equal to 1, 2, 3 etc. Despite this difficulty, the experiments by Fölling *et al* [105] clearly show that commensurability favours localization.

It is not simple to extrapolate such results to the physics of real quantum crystals. One faces the same questions as already mentioned in the introduction: is the crystal commensurate? If yes, is the ratio U/J sufficiently small for superfluidity If not, what is ϵ , the density of vacancies? to occur? Let us consider helium 4 crystals and suppose that they are commensurate. One is tempted to take the activation energy of a vacancy-interstitial pair as an order of magnitude for the interaction energy U. We have seen above that the vacancy energy is approximately 10 K, the interstitial energy is about 20 K and we could take 10 K as an order of magnitude for U. The tunnelling frequency depends exponentially on the height of an energy barrier, which might be of the same order of magnitude. In the case of helium 3 crystals, where the quantum kinetic energy is larger than in helium 4, there is an antiferromagnetic state below 1 mK [109], which is the typical magnitude for the exchange energy. If the exchange energy in helium 4 is at most 1 mK, then it seems to us that helium 4 crystals are far inside the localized state region because U/J is very large.

If helium 4 crystals are incommensurate, it does not seem likely that the vacancy density can be large enough to explain a superfluid transition around 100 mK [34].

6.2. Hysteresis and critical velocity

Aoki et al have studied the NCRIF of the same solid sample at two different frequencies [60]. They use a clever design for the TO, with two masses (one without helium, and one with a cylindrical solid helium sample inside). This system has two resonant modes: at $f_1 = 495.8$ Hz when the two masses move in phase, and at $f_2 = 1172.8$ Hz when they move 180° out of phase. NCRI is observed, with usual features: $T_0 \simeq$ 150 mK and a NCRIF of 0.1%. Below 35 mK, the NCRIF is independent of temperature and frequency; above 35 mK, the low frequency mode at f_1 shows a lower NCRIF (around half that at f_2) and a narrower dissipation peak, 4 mK lower than that for f_2 . Aoki *et al* have also added two interesting pieces of information. First, the dependence of the NCRIF on oscillation amplitude described in section 2 had previously been ascribed to the existence of a critical velocity, by analogy with superfluid helium. However, one may wonder if there is rather a critical displacement or acceleration. In particular, for a rim velocity around 10 μ m s⁻¹ and a frequency of 1 kHz, the rim displacement is only 1.6 nm. Aoki et al have found that the same curve is obtained for the two modes when the NCRIF is plotted against velocity, and not when plotted against displacement or acceleration, confirming the interpretation



Figure 15. Hysteresis effects as observed by Aoki et al [60] at 19 mK (left graph) but not at 63 mK (right graph).

involving a critical velocity. Another important finding is the existence of hysteresis (see figure 15). When the TO is cooled to low enough temperature, e.g. 19 mK, at low velocity near 10 μ m s⁻¹, and the oscillation amplitude then increased, the NCRIF remains constant up to 800 $\mu m s^{-1}$ instead of vanishing; if this is a transient effect, its time constant for decay is estimated to exceed 100 h. If the TO is oscillated at any velocity below 610 μ m s⁻¹ at 300 mK before cooling to 19 mK, the velocity dependence of the NCRIF shows a large hysteresis: when the velocity decreases, the NCRIF increases and saturates below 15 μ m s⁻¹; from this situation, if the velocity is increased, the NCRIF remains constant up to 800 μ m s⁻¹. The two modes exhibit the same behaviour. On the other hand, there is no hysteresis at high enough temperature; the transition between the two regimes is around 40 mK. The observed history dependence is analogous to the effects of 'zero field' or 'field' cooling in superconductors and magnetic materials [110].

Similar results have been obtained by Clark, Maynard and Chan [20], whose systematic study shows the existence of many metastable NCRIF available to the system. A precise interpretation remains to be made, probably in terms of vortex pinning, or in terms of the dynamics of a vortex tangle [82], or ³He impurity pinning either on vortices or on dislocations. It should be eventually noticed that Clark's study [20] shows that, for some samples, the NCRIF keeps increasing down to velocities of order 1 μ m s⁻¹, which correspond to less than one circulation quantum. This is an obvious difficulty in the vortex scenario. Finally, possible effects of a dc rotation on the measured NCRIF have not yet been observed with a rotating cryostat, despite the efforts by Penzev *et al* [58].

6.3. Dissipation peak

Instead of a supersolid interpretation of the NCRI effect, Nussinov *et al* [111] propose that the observed period drop is a natural consequence of the associated dissipation peak: the connection is made in the frame of linear response theory, using the Kramers–Krönig relations. As for the origin of the dissipation peak, they propose that a small liquid-like component transforms into a glass at low temperature; the dissipation reaches a maximum when the equilibration time *s* of this component becomes the inverse of the TO pulsation. Nussinov *et al* are able to give an overall satisfactory fit of the data from Rittner and Reppy [57]: they set several adjustable parameters to reproduce the dissipation peak, and then need one more to explain the temperature dependence of the resonant period. However, Aoki *et al* [60] note that their measurements at two different frequencies (see section 6.2) cannot be both reproduced with the simple form $s = s_0 \exp[\Delta/(k_BT)]$.

In an analysis of the relation between the dissipation peak amplitude and the period shift, Huse and Khandker [112] concluded that most samples must be inhomogeneous so that the dissipation peak is broader than it should be in a homogeneous sample.

6.4. X-ray and neutron scattering

As mentioned in section 3.2, the neutron scattering measurements by Blackburn et al [48] show no measurable variation of the lattice parameter in the temperature range 0.14-0.8 K, in agreement with the existence of a large activation energy $E_{\rm vac}$ for vacancies. The neutron scattering measurements by Adams et al [113] showed no variation of the lattice parameter either, but in a smaller temperature domain (0.07-0.4 K). Both experiments have looked for a possible effect of a superfluid transition on the mean square displacement or kinetic energy of atoms in their hcp lattice. No variation was found as a function of temperature. In the experiment by Diallo et al [114], neutron scattering was used to look for the existence of a condensate in solid helium, but the result was $n_0 = (-0.10 \pm 1.20)\%$. If the anomalies seen in TO experiments are due to a superfluid transition, it has to be associated with defects which are either not present or not visible in these neutron scattering experiments.

Mulders and his group tried to measure the lattice parameter in a much larger temperature range than Fraass *et al* [45] using x-ray diffraction. He reported [115] difficulties in making this measurement. He observed large fluctuations in the mosaic of polycrystalline helium samples and measured all types of temperature variations of the lattice parameter from one sample to another. According to him, the reproducibility of Fraass' results is questionable. We hope that this problem can be solved in the near future. A new experimental determination of the activation energy of vacancies would certainly be useful. We finally note that Ye [116] has calculated a phase diagram for the supersolid phase and predicted that signatures of supersolidity should be observable in x-ray measurements. However, he has taken an equilibrium line for a spinodal line when considering that the roton gap energy vanishes at the melting pressure $P_{\rm m}$, and this is incorrect: the roton gap is non-zero at the melting pressure [121].

6.5. Phase coherence and macroscopic flow

As mentioned in section 2, the evidence for a macroscopic phase coherence in solid helium samples is a little weak. One needs to repeat the experiment of Kim and Chan [10] with blocked and unblocked cells having the same thickness, diameter and height. One also needs to vary the ratio of their thickness to their diameter and compare the ratio of NCRIF to the calculations by Fetter [19] and Mueller [21] of irrotational flow in an annular geometry. If a poor agreement is found with theory, one needs to consider mechanical explanations since the presence of a barrier across the annulus should change the stress field applied to the sample. This is a very important test for many theories which assume macroscopic phase coherence.

Furthermore, there is no strong evidence yet that a dc mass superflow can exist through solid helium at low temperature. We note that Josserand, Pomeau and Rica [117] developed a model based on the Gross–Pitaevskii equation which shows NCRI but apparently no dc superflow. Coming back to experiments, Sasaki's experiment [55] needs to be improved in order to demonstrate the existence of dc superflow along grain boundaries (see section 4.2). On the other hand, pressure gauges apparently work with a good accuracy if the solid sample geometry is sufficiently open. There must be a crossover in size for mass flow to take place, and possibly also conditions on stress gradients or pinning of defects on walls. All these effects should be explored further.

Finally, there is no experimental evidence of phase coherence along dislocation lines and the predictions by Boninsegni *et al* [83] also call for experimental checks.

6.6. Pressure dependence

As shown in figure 4 and briefly mentioned in section 2, Kim and Chan [25] have measured a non-monotonic pressure dependence of the NCRIF in samples grown at constant volume. From the melting pressure $P_m = 25$ bar to about 55 bar, the NCRIF increases and it decreases then up to the highest pressure at which it was measured (135 bar). Since it is generally accepted that the activation energy of vacancies E_{vac} is an increasing function of P, and since energy barriers involved in tunnelling from site to site are likely to be also increasing with P, Kim's results are puzzling. One possible interpretation is that crystals grown at constant volume and at high pressure have a larger density of dislocations or grain boundaries. As for the decrease at even larger pressure, if one assumes that the TO anomalies are related somehow to the superfluidity of dislocations or grain boundaries, it could be due to this superfluidity vanishing at high pressure. It is well known that the superfluid transition temperature T_{λ} decreases with *P* because the roton minimum, which is a sign of local order in the liquid, also decreases. It has been predicted that superfluidity should disappear in metastable liquid helium when the roton energy goes to zero. The calculation by Vranjes et al [121] shows that this gap is not yet zero but strongly reduced at 300 bar. The possible vanishing of superfluidity at high pressure is currently under study [33, 118–122]. It is also possible that, as P increases, the dynamics of dislocation lines or grain boundaries slows down. The pressure dependence of measured quantities is obviously an important check for any model.

6.7. Hydrogen

At an early stage in their series of experiments, Chan and his group reported preliminary results on solid molecular hydrogen (H₂). They had observed a decrease in period, similar to the one in helium, although smaller in amplitude [123, 124]. However, a control experiment with a cell blocked by a barrier showed the same effect. Chan *et al* concluded that hydrogen was apparently *not* supersolid and that the period change could have another explanation, perhaps associated with *ortho–para* conversion [125] and the corresponding phase transition. It would be interesting to clarify the origin of the effect in hydrogen.

7. Perspectives

As announced in the introduction, we believe that the whole issue of supersolidity is not yet understood. Helium crystals show a series of intriguing properties whose existence is firmly established but which still need a clear interpretation. From the theoretical side, a lot of progress has been made in the last few years but there remain controversies on some fundamental questions and, more importantly, none of the existing models seem to be able to make quantitative predictions for all the observed properties. A particularly important challenge is to describe the non-classical rotation properties of torsional oscillators and the change in elastic properties observed by Day and Beamish in a unified and quantitative model. For this one needs to better understand interactions between vacancies, fluctuations and mobility of dislocations and grain boundary, the nature and dynamics of nodes in networks of dislocations or grain boundaries and also ³He adsorption on all these defects. In a comment about Day's experiments, Huse and Dorsey [126] consider the possibility that the changes in elastic properties could be the primary effect with superfluidity a side effect and, once more, this needs to be further checked.

Now that the importance of disorder has been recognized, one needs to make measurements on helium samples with known disorder. The quenched samples of Rittner and Reppy on the one hand, and the single crystals grown by Clark *et al* on the other hand, obviously have a different disorder and their properties do not necessarily need to be described within the same model. The Vycor data fall very far away from all other data on the graph drawn by Rittner and Reppy (figure 4) and this supports the idea that the associated mechanism is different from the one in bulk samples. The measurement of densities of dislocations or grain boundaries is a challenge in itself. Moreover, it would need to be combined with high sensitivity measurements of specific heat, torsional oscillator properties, elastic moduli, mass flow etc.

As we have noted, the evidence for the existence of a macroscopic coherence in solid helium samples is weak and we hope that this crucial point is carefully studied with various shapes of blocked and unblocked cells in the near future. One would also like to obtain strong experimental evidence that grain boundaries are superfluid as predicted, also the core of dislocations. The mobility of these objects is an interesting problem in itself, as well as the energy and dynamics of nodes in dislocation networks, contact lines of grain boundary, ³He adsorption and mobility on these defects. Experiments in hydrogen and its isotopes might be useful to repeat as well.

There must be many other questions to solve, both experimental and theoretical ones, and we apologize for having certainly not mentioned all of the ideas on supersolidity which have been published yet. In fact, we would be very interested in discovering new ideas or questions on this fascinating subject.

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