Solid He4: A Case Study in Supersolidity

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Abstract

Solid He4 currently is the strongest candidate for realizing the proposed "supersolid" state of matter. Recent experimental results, however, have served to significantly complicate the original picture of solid He4 as a supersolid. In an attempt to get a handle on what has turned out to be a broad and rapidly changing field, I will present the original theoretical understanding as laid out by Andreev and Lifshitz, Chester, and Leggett, summarize the experimental data since Kim and Chan's discovery of non-classical rotational inertia, and then describe how modern theoretical work has developed to accommodate some unusual discoveries.

1 Historical Introduction

The existence and general properties of a supersolid state of matter were proposed about forty years ago, generally credited to Andreev and Lifshitz' paper in 1969, and to Chester's in 1970[1, 2]. In it, they proposed a novel phase of matter in which a system, while ostensibly solid, could also exhibit superfluid phenomenon due to quantum effects. Though the superfluid state had been realized before in He4, this supersolid state represented an entirely different beast. Understandably, quite a bit of excitement was generated in 2004 when Kim and Chan observed non-classical rotational inertia in solid He4 (a necessary condition for supersolidity), and so the hunt began to characterize this novel state and understand its underlying mechanism. The enthusiasm is justified, as solid He4 seems to be the only possible realization of a supersolid state at this time, although such a state could be produced in optical lattices [17].

As recent experimental and theoretical studies have discovered, He4 turned out to be full of surprises, and it is currently agreed that if supersolidity exists in He4, its mechanism is significantly different from the early theories of bulk supersolids. However, these initial papers are indispensable in framing the question of "what is a supersolid". Accordingly, I will highlight important points from the work of Andreev and Lifshitz, Chester, and Leggett. Over the course of this, the properties of the expected supersolid state, along with experimental tests for these properties, should become readily apparent.

1.1 Andreev and Lifshitz

In "A Quantum Theory of Defects in Crystals" [1], Andreev and Lifshitz start off considering a crystal consisting of atoms with zero-point oscillations that are large, but not so large as to be comparable to the lattice spacing. This is primarily to ensure that we can still talk of atoms being roughly localized to crystalline lattice sites. They then consider a defect in this crystal–either a vacancy, interstitial atom, or later, an impurity atom. Though classically, this defect can diffuse throughout a crystal by thermal excitations, quantum effects also allow this defect to tunnel to neighboring sites. Treating this defect as its own type of particle, Landau and Lifshitz state that one can label the quantum states of this defect with a quasimomentum vector $\mathbf{k}$ and a dispersion relation $\epsilon(\mathbf{k})$. Due to the time-energy uncertainty relation, the energy uncertainty $\Delta\epsilon$ should be proportional to the tunneling probability. Thus, we should expect the dispersion relation to be centered around some value with a bandwidth given by $\Delta\epsilon$. But notice what has happened–they have taken a defect with a finite probability of tunneling, and have managed to produce a series of states with a well-defined quasimomentum and dispersion relation. These are nothing but quasiparticles–"defectons" as Andreev and Lifshitz call them.

An important point is that these defects can obey either Fermi-Dirac or Bose-Einstein. This will depend on the statistics of the crystal's constituent atoms, and on the statistics of the defect involved. They show that vacancies obey the statistics of the atoms in the crystal. For interstitials, the defect obeys the statistics of the interstitial atom. For substitutions on lattice site, the defect is a boson if the substituted atom and the crystal's constituent atoms have the same statistics, and is a fermion in the opposite case. Thus, we would expect that for He4, vacancies and interstitial He4 atoms would be realized as bosonic defectons at suitably low temperatures. He3 impurities however, would form fermionic defectons.

Assuming that the bosonic defectons are suitably delocalized and assuming that they are present in equilibrium, they will form a Bose-gas inside a crystal. Andreev and Lifshitz deal with these two criterion individually. Delocalization of defectons depends on interactions with other crystalline excitations (phonons for example) and with other defectons. Let n be the concentration of defectons per unit volume, a be the distance between lattice sites in our crystal, m be the mass of a constituent atom in our crystal, and c the speed of light. Then assuming the temperature is low enough to yield non-localized defects, the defects will be non-localized if $na^3 \ll (\Delta\epsilon/mc^2)^{3/4}$ holds.

That defectons will be present at suitable equilibrium conditions is a more subtle point. To summarize, Andreev and Lifshitz argue that for suitable temperatures and pressures, a system can actually minimize its free energy by having defectons present (of the vacancy or interstitial type). Not only this, but in equilibrium, bosonic defectons will condense to form a BEC in the $\mathbf{k} = 0$ state. They predict a first-order phase transition at non-zero temperature from a state of negligible defecton densities to macroscopically large defecton densities in a condensate.

This prediction is perhaps the most important part of their paper–that Bose-Einstein condensation of defects can occur in a solid. As such, they assert that a superfluid state should exist within the crystal lattice, composed of these defectons. Though the condensation is among the defectons, when vacancies or interstitials move through a lattice, there is a corresponding mass transport associated with the constituent atoms, much akin to hole currents in semiconductors. They also suggest the startling possibility that a solid exhibiting this superfluid mass-transport phenomena should be "... able to flow through a capillary in a gravitational field". More on this in the experiment section of this paper.

There are two important points to note about this theory, particularly when it comes to He4. The first is that it predicts superfluid flow through the bulk of a quantum solid. Secondly, the mechanism is crucially dependent on the presence of zero point defects, both at zero temperature and finite temperature. In other words, this theory applies to "incommensurate solids", or solids without an integer number of atoms in a unit cell. We will see later that recent theoretical and experimental work largely contradicts these two points.

1.2 Chester and Leggett

It is fascinating to note that in 1970, Chester also indicated the possibility of Bose-Einstein condensation in bulk systems characterized by periodic order, but his approach was radically different [2]. Unlike Andreev and Lifshitz, Chester considers approximate forms for many-body bosonic wave functions. He begins by starting with a general bosonic Jastrow wave-function of the form $\psi_N = exp(-\frac{1}{2}\sum_{i\neq j} u(r_{ij}))$, where $u(r_{ij})$ is any pseudopotential that is "... hardcore, is bounded below and has finite range". What he notes is that he can chose the $u(r_{ij})$'s such that the probability distribution associated with this wave function will mimic a classical Gibbs distribution. Since we know that such distributions can represent crystalline systems at suitable temperatures and pressures, he concludes that for low enough temperatures, this wave function will reflect a crystalline state. Invoking a theorem by Reatto, Chester shows that such a system can also form a BEC.

The application of this result to real physical systems fell more into the realm of speculation, but are important nonetheless. He stated that a bosonic Jastrow function should be a good approximation to the states of solid He4, and so there was a pretty good chance of solid He4 exhibiting Bose-Einstein condensation. What would also turn out to be of great importance in the current supersolid He4 debate would be his suggestion that Bose-Einstein condensation in a quantum solid might only be possible when the state of a system includes a macroscopically large number of vacancies. This would later be born up in more rigorous theoretical treatments.

On the heels of Chester's article, Leggett's paper "Can a Solid be a Superfluid?" greatly elaborated on the macroscopic physical characteristics that such a combined solid, superfluid state of matter should exhibit [3]. Most importantly, he predicts that a solid with a superfluid like component should have a rotational inertia that is a little less than its classical value. This "non-classical rotational inertia" (NCRI) arises because a fraction of the mass of the solid essentially decouples and condenses into a superfluid state, which can only acquire angular momentum in discrete quanta [11]. This observation, and his suggestion to rotate a sample of solid He4 to observe this effect, was eventually realized in the 2004 Kim and Chan experiment [4]. It should also be pointed out that Leggett, on the assumption that a supersolid state existed, managed to place an upper bound on the fraction of a solid that can exhibit superfluidity. His original crude estimate put this fraction to be less than ~0.01%, which was *mostly* realized when better estimates became available [3, 7, 13].

As a result of this discussion, we can now see what qualities we expect the "supersolid" state of matter to exhibit. It is an ordered, crystalline solid that also exhibits superfluid-like mass-transport properties on account of Bose-Einstein condensation. The presence of this state can be indicated by the existence of NCRI, or by observing the solid flow through a capillary. Lastly, the presence of BEC indicates that this is a distinct phase of matter, with Andreev and Lifshitz predicting a first-order phase transition into this state below a certain temperature, and above a certain pressure [1].

2 Experimental Work

The picture concerning a possible supersolid mechanism in He4 changed considerably as a result of puzzling experimental evidence. The experimental work is quite extensive (See [5] for a full review), so I will first highlight some important results in favor of a supersolid interpretation, namely the work of Kim and Chan in 2004, and *part* of Reppy in 2008. I will then present some interesting experiments that challenge this interpretation, namely Day and Beamish in 2006, Sasaki in 2006, and Reppy again in 2010.

The first serious experimental evidence for a supersolid state of He4 came with Kim and Chan's 2004 experiments, published in Science and Nature [4]. As their setup is highly typical in the field, I will describe it in some detail. In it, they used a torsional oscillator (fig 1). It consists of a rod attached to a bob, which contains solid He4 grown in Vycor glass, a porous material. Any change in the rotational inertia of the sample is observable by a change in the resonant period of the oscillator, given by $T = 2\pi\sqrt{I/G}$, where G is basically a torsional spring constant, and I is the rotational inertia of the bob.

As is indicated in Figure 1, when the sample was cooled to below ~170mK, a noticeable drop in the in the oscillator period is noticeable, corresponding to a drop in the rotational inertia of the sample. Such a drop was not observed when the cell was empty, or when a cell was similarly prepared with He3. Similar experiments corroborated these findings [7]. They also noted in this paper that the NCRI was very sensitive to the concentrations of He3 present in the sample, adversely being affected with higher concentrations. Henceforth, I will be very careful not to use "supersolid" fraction, as this presupposes that the mechanism behind NCRI is a supersolid state. Instead, I will use "non-classical inertia fraction", henceforth referred to as NCRIF. This is defined as the ratio of the NCRI discrepancy over the classical rotational inertia. If supersolidity exists, this gives the "supersolid fraction".

Since Kim and Chan's initial experiment, their results have been replicated, and further refinements were made. Experiments were made on solid He4 grown in porous gold, and eventually

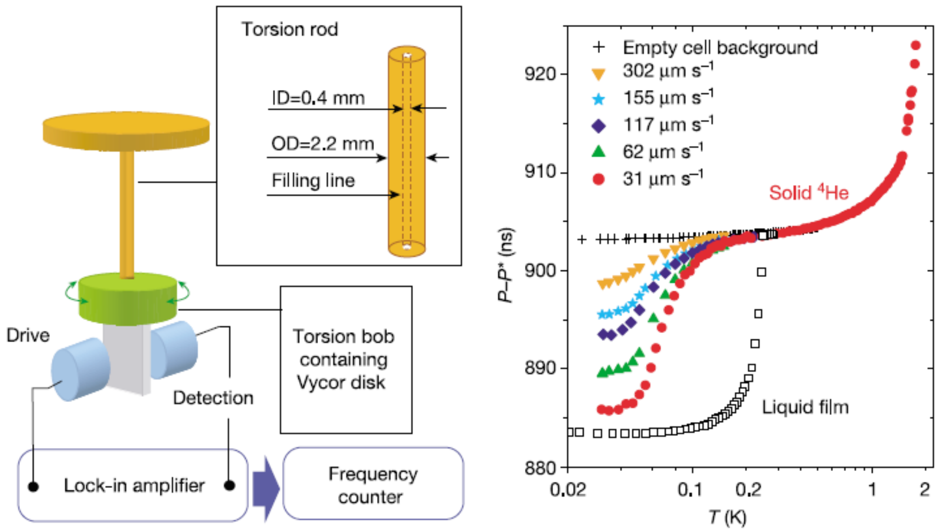

Figure 1: **left** : Torsional oscillator setup, **right** : Difference in oscillator period with respect to temperature. The maximum velocity of the edge of the Vycor disk is shown in the legend, and it corresponds to the amplitude of the oscillations. (Taken from [4])

on bulk solid He4 [5]. Most favorably to a supersolid transition, an experiment was conducted by Reppy (originally by Kim and Chan), where he created an annulus of solid He4 [6]. The twist was that he had placed an impermeable barrier on diametrically opposed sides of the annulus, thereby effectively segmenting it. When he measured the non-classical rotational inertia in his torsional oscillator experiment, he observed that the NCRI disappeared to within experimental errors. This is taken to be a strong indication that the cause of NCRI in solid He4 samples is due to superflow within the solid sample, as such a flow should be interrupted with a discontinuous annulus.

Anomalies began to pile up however. As mentioned before, Leggett derived a rigorous upper bound for the supersolid fraction that depended eminently on the density matrix at microscopic levels [3]. As such it is expected that the supersolid fraction is a quality of a bulk material, not dependent on the macroscopic geometry of the sample. However, Reppy noticed some "puzzling" results in the previously mentioned paper, such as the fact that the superfluid fraction seemed to be dependent on the cell geometry [7]. For larger, "less confined" samples, Reppy found the estimated NCRIF to be ~0.05%, which is considered reasonable in the field and consistent with Leggett's upper bound on the supersolid fraction. However, when using very thin tightly confined annuli, Reppy found the NCRIF to be as high as 20%, which is highly anomalous [6].

Reppy found several more concerning anomalies in other papers. The following are taken from a review article [5]. In 2006 while replicating the Kim and Chan experiments, Reppy and noticed that the NCRI of He4 seemed to depend on how the solid He4 sample was grown. Specifically, by using a process known as "annealing", Reppy was able to grow very pure and highly ordered samples of solid He4. Oddly enough, this seemed to *decrease* the estimated NCRIF. Correspondingly, they observed that when a solid was formed by "quenching", or forming a solid by rapid cooling, that the estimated supersolid fraction actually increased. These experiments indicated that if supersolidity

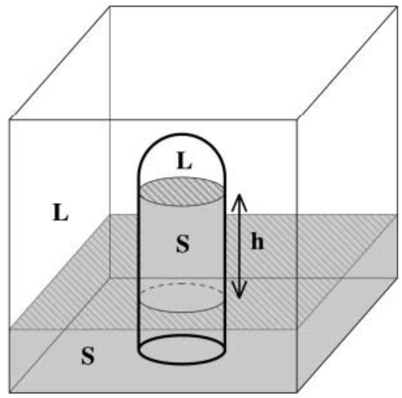

Figure 2: Superfluid apparatus for Sasaki et al. Taken from their paper [10]

does exist, it is dependent on the amount of disorder present in a sample.

Furthermore, in 2010, Reppy looked at the effect of plastic deformation of solid He4 [8]. This was again accomplished using a torsional oscillator. This time, a diaphragm was included so as to be able to be able to apply stresses to solid He4 samples. Based on a disorder-induced supersolidity model, what one would expect to happen would be that stress-induced disorder should produce a noticeable change in the NCRI below the transition temperature. On the other hand, above the transition temperature, a mechanism for superflow shouldn't be present, and so we shouldn't expect a change in the rotational inertia. However, Reppy observed that the largest differences in the rotational inertia of the sample occurred above the transition temperature, with the differences in NCRI between both samples actually converging for temperatures below the transition temperature. This was such an unusual result that Reppy suggested a non-supersolid model for He4 might be in order.

More unusual were the measured shear modulus anomalies below the proposed supersolid transition temperature by Day and Beamish [9]. Intuitively, it is expected that when a solid becomes partially liquid, its shear modulus should drop accordingly. Using a torsional oscillator, and measuring shear with piezoelectric sensors imbedded in the solid He4 sample, Day and Beamish measured a noticeable*increase* in the shear modulus of the solid He4 sample below a transition temperature of about 200mK. They also observed that this anomaly was dependent on He3 concentrations, and suggested that there was a common underlying mechanism for both superflow and the anomalous shear modulus increase.

Lastly, in a particularly interesting experiment by Sasaki et al. [10], they grew solid He4 within and around a test-tube immersed in liquid He4 (fig 2). Note that the highest point of the solid He4 is about a centimeter higher than the rest of the solid He4 in the apparatus. From Andreev and Lifshitz' theory of supersolidity, since we have a gravitational field, the presence of a gravitational field will cause mass-transport of He4 from within the test tube to the surrounding area. This will be manifested by a drop in the solid level within the test tube, and a corresponding increase in the solid level outside the test tube. This process will continue until the solid level inside and outside the tubes are equal. Surprisingly, this phenomenon was not observed in most of the crystal samples they grew. High quality samples with no visible fractures remained in their initial configurations for the duration of the experiment. Their "medium" quality samples, those with a few visible fractures, did however exhibit mass transport, evidenced by a lowering of the solid level within the test tube. Their lowest quality samples ended up reaching an equilibrium height the fastest of all three grades of samples. They ended up concluding that mass-transport was caused by the presence of "grain boundaries", which could support superflow. More on this in the next section.

3 Theoretical Developments

Before delving into an in-depth summary of recent theoretical attempts to understand the anomalous low temperature behavior in solid He4, one needs to appreciate what is meant by "off-diagonal long-range order", or ODLRO [11, 12]. The concept of off-diagonal long-range order has emerged as a clean, necessary condition for establishing the existence of Bose-Einstein condensation [17]. To understand off-diagonal long-range order, one looks at the function known as the "single-particle density matrix". It is defined as follows: $\rho(\mathbf{r},\mathbf{r}') = \langle \psi^\dagger(\mathbf{r})\psi(\mathbf{r}')\rangle$, where $\psi(\mathbf{r}')$ annihilates a particle at $\mathbf{r}'$, $\psi^\dagger(\mathbf{r})$ creates a particle at $\mathbf{r}'$, and the brackets indicate an ensemble average, formed by tracing over this operator times the N-particle density matrix of our system [11]. Now, a system is said to have off-diagonal long range order if in the thermodynamic limit, this quantity tends to a non-zero constant in the limit as $|\mathbf{r}-\mathbf{r}'| \to \infty$. Intuitively, this indicated the presence of correlations between particles separated by an arbitrarily large distance.

As there is a fundamental relationship between ODLRO and the presence of Bose-Einstein condensation, this concept has been of fundamental importance in recent discussions on BEC's and supersolidity. Thus, if ODLRO is present, one has a strong indication that a BEC state should exist–whereas if ODLRO is not, then a BEC state certainly shouldn't exist. Note that this idea was not explicit in the papers mentioned at the beginning of this paper.

3.1 Against the Bulk Supersolid State

As mentioned before, the Andreev-Lifshitz model of supersolidity requires the existence of zero point vacancies and interstitials. Shortly after the original Kim and Chan experiments, various computational and theoretical groups set about trying to establish two things. First, whether supersolidity was possible in a quantum crystal without vacancies or interstitials. Secondly, to establish whether or not He4 has the required number of vacancies in the temperature range below the observed critical temperature in experiments.

Prokof'ev in 2005 managed to show analytically that vacancies and interstitials were necessary in order for there to be supersolidity in the bulk of a crystal [14]. He accomplished this by topological considerations of a ground-state bosonic wave function, and also by path-integral arguments. Unfortunately, the activation energy for defects in He4 was calculated to be approximately 14K in Monte Carlo simulations performed by Pederiva et al. (taken from the referenced Ceperley paper), Ceperley would follow up with this in 2004 to calculate the activation energy for an interstitial, which was about 48K [13]. As such, it was shown that contrary to Andreev and Lifshitz, He4 does not have a macroscopically large number of vacancies or interstitials in the temperature range experimentalists were looking at, and so the mechanism of supersolidity in He4 would have to be more complicated.

Ceperley also attacked the problem more directly with a 2006 paper where he calculated the off-diagonal single particle density matrix elements for the ground state of He4, again using path-integral Monte Carlo methods [12]. He showed that rather than going to some finite constant, this quantity goes to zero in the thermodynamic limit and for large particle separations, indicating that off-diagonal long range order was not present. One can therefore conclude that Bose-Einstein condensation is probably not present in bulk, pure He4 at the temperature and pressure ranges experimentalists were probing. This has forced theoretical considerations of the underlying mechanism of proposed supersolidity towards disordered systems.

3.2 Superglass

A superglass refers to a solid state of matter lacking crystalline order yet possessing off-diagonal long-range order, thereby allowing Bose-Einstein condensation and superfluidity in the bulk. This possibility was first shown in a path-integral Monte Carlo simulation by Boninsegni et al. in 2006 [15]. They took between 200 and 800 particles and in a sense "quenched" them down to 0.2K in the simulation. An important caveat is that this is a path-integral Monte Carlo simulation, so these dynamics are somewhat artificial. Regardless, they found a stable system configuration akin to a glass. More surprisingly, this configuration possessed ODLRO in the simulations. This proposed state allows for superflow, and so could explain the NCRI observed. It also gets around the problems associated with pure He4 not being a supersolid.

Recent developments regarding this theory include an analytical treatment of this superglass phase of matter in 2008 [22]. As Chan pointed out though, a glassy state should be observable by well known experimental methods, such as X-Ray diffraction [16]. So far, this has not been corroborated.

3.3 Grain Boundaries and Defects

The Sasaki et al. experiment discussed previously forced theorists to consider the role that structural defects had in superfluid behaviors exhibited by solid He4, particularly grain boundaries. The main types of defects considered were grain boundaries, edge dislocations, and screw dislocations [7]. For now, I will only consider the first two. A grain boundary is defined as an interface between two crystal grains. An edge dislocation occurs when an extra plane of atoms is inserted (or removed) partially into a crystal lattice [17]. The theoretical underpinning for subsequent theories comes from the fact that though pure solid He4 does not possess a significant fraction of vacancies at low temperatures, vacancies can form in and around defects, and so there is a possibility of having a superfluid or supersolid state confined to these regions [7].

Pollet et al. dealt with the possible superfluidity of grain boundaries and defects in 2007 and 2008 respectively [18, 19]. Concerning the first, using an extensive path-integral Monte Carlo simulation, they found that there is a nonzero condensate fraction along a grain boundary. Thus, one can conclude that there will be a superflow. They stress that this does not hold for all grain boundaries, and seems to be dependent on the relative angles between grain boundaries, as well as on unusual configurations of multiple grain boundaries. These special types of grain boundaries, their simulations indicate, will not allow superflow. In their 2008 paper, they show that the activation energy for vacancies in solid He4 drops from the previously mentioned value of 14K to close to 0K when suitable stresses are present, which they are likely to be around defects. Thus, one can strongly speculate that in and around defects, one will have a superfluid flow of vacancies, akin to the Andreev-Lifshitz theory.

Beyond explaining how the presence of crystalline defects can give rise to superflow, Aleinikava and others have invoked defects to explain the anomalous increase in shear modulus below the proposed supersolid transition temperature, as previously described in Day and Beamish's work

[20]. The quantum mechanical treatment they give edge dislocations can be summarized with the following points. They show that at zero temperature, edge dislocations straighten and are pinned to He3 impurities, a state known as a "quantum smooth" state. This edge dislocation can become excited and deviate from this quantum smooth state, but there is a gap associated with this. Until a high enough temperature, the defect lines will remain rigidly pinned, corresponding to a high stress modulus for the sample. With a high enough temperature though, thermal excitations can overcome this gap, and so the defect lines will deviate away from the smooth state. Specifically, the dislocation line can develop "kinks", as well as acquire some mobility [7]. As these dislocation lines are not nearly as rigid as before, this corresponds to a drop in the shear modulus. If one grants them the assumption that superfluidity will be adversely affected in the excited states of the edge dislocations, then this theory manages to tie the NCRIF and shear modulus together.

3.4 Non-Superfluid Mechanism

Iwasa in 2010 dispensed with superfluidity altogether and dealt instead with the role that edge dislocations have in affecting the rotational inertia of solid He4 [21]. In this theory, he was able to attribute not only the stiffening of the shear modulus to the motion and pinning of defects on He3 impurities, but he was also able to attribute the non-classical rotational inertia to this same mechanism. This was invoked by Reppy to justify the anomalous findings in his 2010 study, but he is careful to point out that this theory does not take into account the bosonic nature of solid He4, and so a more thorough treatment might be required.

4 Future Work and Conclusion

Obviously, that supersolidity exists in He4 and what its mechanism would be are still unresolved debates. As such, there is a wide range of possible research directions. However, based on my readings, I can suggest a few theoretical and experimental avenues that I believe are worth looking into.

First, the 2010 Reppy experiment should be replicated. It is a highly puzzling set of findings that could change the supersolid debate landscape significantly. As such, it warrants a second look.

Second, the Iwasa theory should be experimentally tested. Fortunately, Iwasa himself provides a simple test of his theory. Instead of performing a blocked annulus experiment described earlier in this paper, one should perform a partially blocked annulus experiment on solid He4 [21]. This should not affect superflow in the annulus if it exists, but as Iwasa points out, it should have an adverse effect on the propagation of defects. Additionally, Reppy points out that the assumed defect concentration in solid He4 seems unreasonably high in Iwasa's model [8]. Either quantitative measures of edge defect densities in current samples, or a means of growing ultra pure solid crystals of He4 should be looked into to gauge whether or not Iwasa's theory is reasonable.

Lastly, though I found the Aleinikava et al. model extremely appealing, the assumption that superflow and the adverse affects kinks in edge defects have on them should be verified. As path-integral Monte Carlo methods have been successfully applied to problems of this sort, particularly by Prokof'ev and Boninsegni, one could attempt a detailed calculation of how superflow can be affected by different edge dislocation geometries.

Solid He4 represents the closest realization to a highly novel and counter-intuitive state of supersolidity to date. Even if supersolidity does not turn out to be realized, solid He4 continues to elude description, and on those grounds alone warrants further study. More practically, however, solid He4 represents a strongly interacting system that manifests unusual quantum phenomenon macroscopically. A continued pursuit of the anomalous effects exhibited by He4 may very well prove fruitful in our attempt to understand other strongly interacting systems, as detailed studies of superfluid He3 and He4 have already done.

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