Superfluidity in Solid Helium-4

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Superfluidity in solid Helium-4 was predicted in 1969 and has been experimentally realized in 2004. In this essay, I will introduce the discovery of the intriguing emergent state, supersolid in Helium-4, and its recent experimental progress. In addition, since some of the details of the experiments are not yet well understood, theoretical arguments and comments about this quantum phenomenon will also be discussed.

1 Introduction

Liquid ^{4}He undergoes Bose-Einstein condensation to enter a superfluid state without viscosity when it is cooled below a critical temperature of 2.176K. In 1995, similar superfluidity behavior was also realized by the Bose-Einstein condensation of alkali atoms in gaseous phase [1,2]. An interesting question comes out: "Does superfluidity exist in the solid phase?" As a matter of fact, the existence of supersolid- the superfluidity in solid- was predicted by Russian physicists Alexander Andreev and Ilya Liftshitz in 1969 [3]. It was suggested that the supersolid state emerged from the condensation of atomic vacancies, which behave as a coherent entity moving throughout the solid like a superfluid. In addition, when the normal solid enters the supersolid state, it acquires the non-classical rotational inertia, where the vacancies decoupled from the normal solid [4].

However, not many elements are predicted to have this weird quantum state. Only weakly-bound element like ^{4}He, which solidifies only under extremely high pressure and low temperature, could become a supersolid. Due to the quantum-mechanical zero-point energy, even being cooled to near zero temperature, the He atoms still have vacancies, which probably play a crucial role in the formation of superfluid in solid.

2 Primitive Experiment

Because of this specific property, experimental physicists tried very hard to find if the superfluidity presents in the solid ^{4}He. In the beginning of 2004, Eun-Seong Kim and Moses Chan of Pennsylvania State University observed this counterintuitive phenomenon [5]. The experiment searching for the supersolid was conducted by a torsional oscillator whose resonant period is given by $2\pi\sqrt{I/G}$, where I is the rotational moment of inertia of the torsion cell with ^{4}He , and G is the torsional spring constant of the Be-Cu torsion rod, as shown in Fig. 1. In the torsion rod there is a small channel which allows the introduction and pressurization of ^{4}He to become solid. Beside the torsion cell a pair of electrodes is used to drive the cell and keep it at resonance. If the superfluid in the solid ^{4}He occurs, the moment of inertia and oscillation period decrease. Kim and Chan put the ^{4}He into a porous Vycor glass disk whose channel size is about 7 nm in diameter and 30 nm long. It was believed that the solid ^{4}He grown inside such a porous Vycor glass is likely to be heavily populated with lattice vacancies. When further cooled and pressurized the solid ^{4}He in Vycor glass, it was found that there is a significant drop of period around 175 mK (Fig. 2.). This important information implies that around 175 mK some fraction of confined solid ^{4}He starts to undergo a phase transition to become supersolid. A noticeable point is that even though the amplitude and velocity of the rotating disk

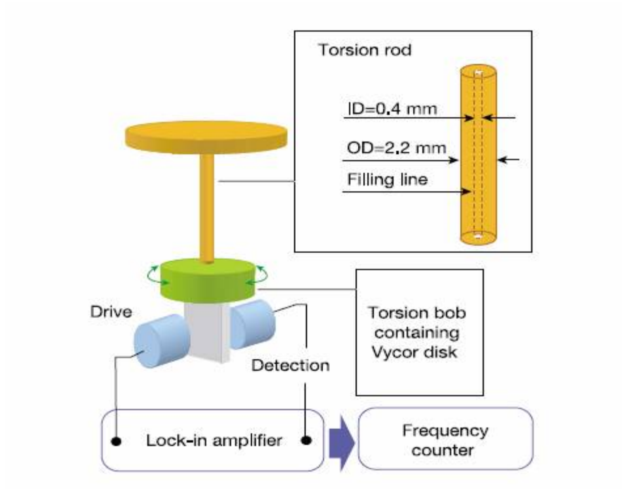

FIG.1. The torsional oscillator used to search for the supersolid. ^{4}He is put in the Vycor disk with diameter of 15 mm and thickness of 4 mm. A pair of cylindrical electrodes is used to drive and detect the oscillation of the Vycor disk. Since the mechanical Q of the oscillator is 10^6 at low temperature, the resonant period can be measured precisely to 0.2 ns [5].

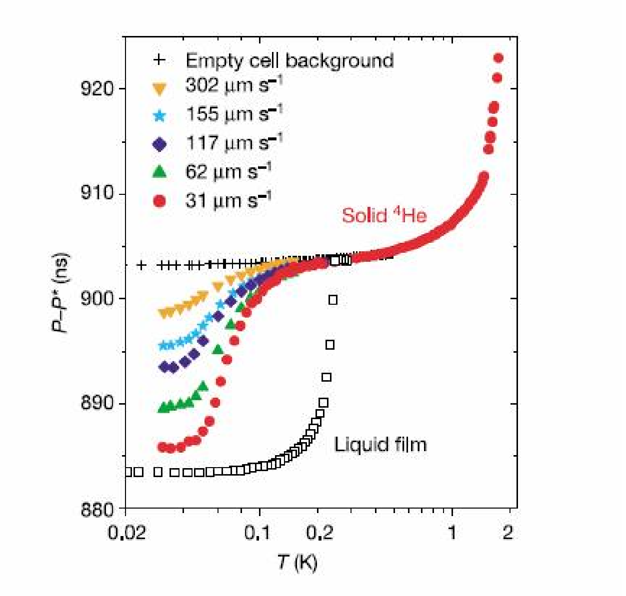

FIG. 2. The measured resonant period as a function of temperature for different rotating velocities of the solid ^{4}He samples confined in the Vycor disk. A common onset of period drop around 175 mK was observed, which is probably a signature of the supersolid phase transition. Notice that the period curve of thin liquid film adsorbed on the walls of internal space of Vycor is remarkably different from those of solid ^{4}He samples [5].

strongly determine the resonant period, the transition temperature around 175 mK does not rely on the rim velocity. On the other hand, in order to exclude non-supersolid mechanisms contributing to the observed phenomenon, Chan also made the same measurements on pure solid ^{3}He and solid ^{4}He with 10, 30, 100, 1000 and 10,000 p.p.m. of ^{3}He. The results are consistent with their expectation that the addition of fermionic ^{3}He will ruin the supersolid. The higher concentration of ^{3}He is added, the higher transition temperature or even no transition occurs. However, since it is still questionable that whether the solidification in Vycor glass is complete or not, it could be possible that the observed non-classical rotational inertia comes from a thin liquid film of ^{4}He that had been trapped in the nanoporous glass disk. In any case, as a surprising experimental achievement which had been predicted for few decades but not realized, even though this "probable observation" of supersolid is incipient and immature as well as the microscopic origin is not understood, this result really opens an intriguing and promising filed again and excites many other devoted physicists to investigate this phenomenon.

3 Self-confirmed Experiment

Later 2004, Kim and Chan repeated their experiment using a bulk solid ^{4}He sample confined to a revised annular channel in a torsion cell [6]. They measured the non-classical rotational inertia fraction as a function of temperature for 17 solid samples as well as observed the drop of the resonant oscillation period. It was estimated that as the cell was cooled down to around 230 mK, the solid ^{4}He entered a superfluid state. Furthermore the phase diagram of normal solid and supersolid ^{4}He was also mapped out from 26 to 66 bars, as shown in Fig.3. From these results Kim

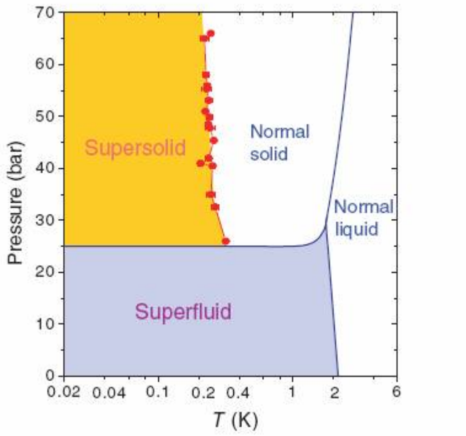

FIG. 3. Phase diagram of the liquid and solid ^{4}He. A transition from normal solid to supersolid is also mapped out [6].

and Chan confirmed their previous experiment of the realization of superfluid in solid ^{4}He and claimed that this phenomenon is intrinsic and not the result of confinement in porous or any particular medium.

4 Further Confirmation from Allies and the Role of Disorder and Grain Boundary

After Kim and Chan reported they discovered the superfluidity in solid ^{4}He, there is no other experimental group endorsing this intriguing result until 2006. Among which, Rittner and Reppy of Cornell University confirmed this phenomenon by using a similar technique and apparatus that they sealed the solid ^{4}He in a torsion cell and cooled to measure the drop of resonant period [7]. A noticeable point is that a heater and a thermometer were mounted on the cell which can heat up the sample rather than only cooling. After repeated measurement, the onset of non-classical rotational inertia of solid ^{4}He was identified below 250 mK. Furthermore, the Cornell group also investigated the effect of annealing on the supersolid signal. By heating up the solid ^{4}He sample to its melting point for few hours and then slowly cooling down again, the supersolid signal can be reduced or even can disappear by the fully annealing (Fig. 4). Since the purpose of annealing is to reorganize the crystal and reduce its imperfection,

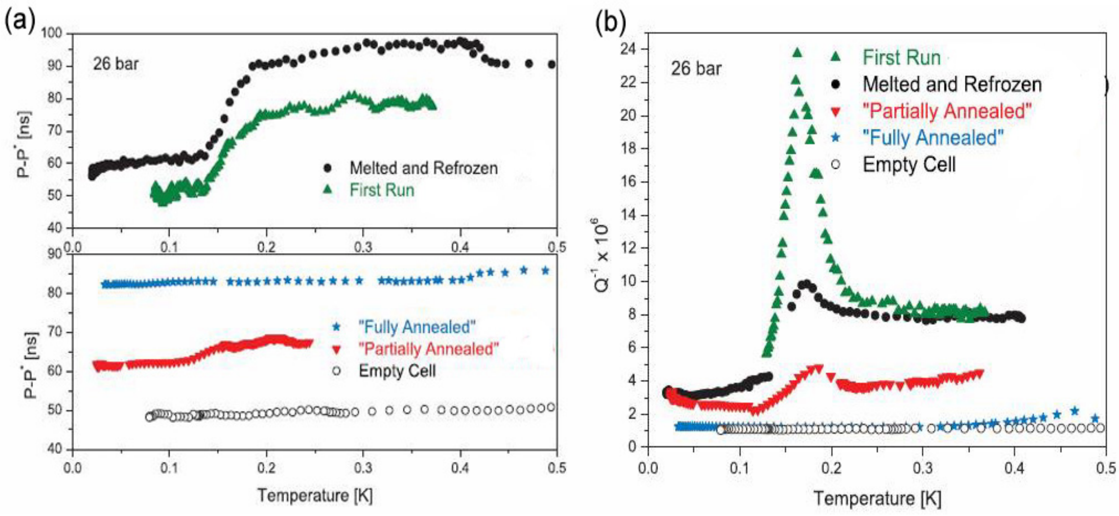

FIG. 4. (a) Resonant period as a function of temperature for samples with different preparations. Notice that the behavior of period for the first run is remarkably different from the partially and fully annealed samples. (b) Dissipation as a function of temperature for samples with different preparations. The annealing effect gradually reduces the dissipation peak. (For the detail, see [7])

the observed mitigation of period drop and dissipation data as a function of temperature suggested that the superfluidity in solid is closely related to the imperfections such as vacancies and disorders of the crystal structure as well as that the supersolid is not a universal property of ^{4}He and can be diminished or even removed by annealing. The absence of superfluidity in solid ^{4}He was also consistent with the result done by Ceperley's path-integral-Monte-Carlo calculations that an ideal hexagonal-close-packaged ^{4}He does not support the off-diagonal-long-range order [8].

To further study the disorder effect on the supersolid, the Cornell group rapidly froze the normal liquid ^{4}He in order to form a highly disordered solid sample, in which the whole process of freezing and cooling is finished within two minutes [9]. Interestingly a noticeably large drop of period, about 20 % - 30 % of entire solid moment of inertia, was observed. On the other hand, when the sample was annealed slowly for 14 hours to form a highly ordered structure, there is only 6 % of the ^{4}He becoming supersolid. These results strongly suggested that the disorder plays an important role in supersolid as well as implied the theory of Andreev and Liftshitz, which predicts the strength of supersolid is from zero-point vacancies alone, is not a complete story as both experimentalists and theorists suspected in recent years.

On the other hand, some groups also suggested the grain boundaries of superfluid could be the possible mechanisms causing supersolid signals [10,11]. Svistunov of University of Massachusetts even claimed that based on first-principle calculation there are at least two supersolid phases. One of them occurs in the grain boundaries of superfluid, which are about few atoms layers separating different crystal orientation regions; the other is the glass phase of superfluid where the atoms form a metastable superglass state [12]. However, Chan excluded the possibility of grain boundary by repeating their experiments with single crystals of soild ^{4}He which also show supersolid signals and suggested that the origin of supersolid could be from the dislocations within crystals [13]. This suggestion was quickly backed up by a simulation of a screw dislocation in the microscopic ^{4}He crystal [14]. Svistunov's group found that the core of screw dislocation behaves as a tube and some atoms could freely flow through it when the temperature approaches absolute zero. This suggests that in this situation the system is essentially like a superfluid and also provides specific signatures that could be experimentally examined.

5 Thermodynamic Signature of Phase Transition

Even though more and more research groups had observed similar phenomena of supersolid transition, there is still no theoretical consensus on the mechanism and nature of supersolid. In order to provide a thermodynamic characteristic of phase transition from normal solid to supersolid, Chan's group at Pennsylvania State University measured the heat capacity of the solid ^{4}He with dilute ^{3}He of 1 p.p.b., 0.3 p.p.m., 10 p.p.m. and 30 p.p.m [15]. When cooling down the samples, a noticeably excess heat capacity besides Debye term (T^3) at low temperature was observed, in particular for the cases of 10 p.p.m. and 30 p.p.m. ^{3}He impurities. Since it was suggested that the presence of the non-classical rotational inertia is due to the formation of glassy state of solid helium, whose heat capacity has a linear temperature-dependent signature, the temperature dependence of specific heat was plotted in order to test the possible evidence of glassy state. Figure 5 shows the specific heat divided by T (C_n/T) as a function of T^2 and indicates there is a non-zero intercept at y-axis for both 10 p.p.m. and 30 p.p.m. ^{3}He impurities. This suggests that, in addition to Debye T^3 term, there could be a component linearly scaling with T. In other words, the specific heat might have the form: $C_n \sim aT + bT^3$, where a is the y-intercept and b is the slope in the C_n/T against T^2 plot. However, according to the measurement of linear term of specific heat by other group, this correction term would be reduced by annealing of the sample [16]. Moreover Chan's group failed to find any positive evidence, like the hysteresis or time-dependent signature, for the presence of glassy state. Then they further ruled out the glassy state possibility by

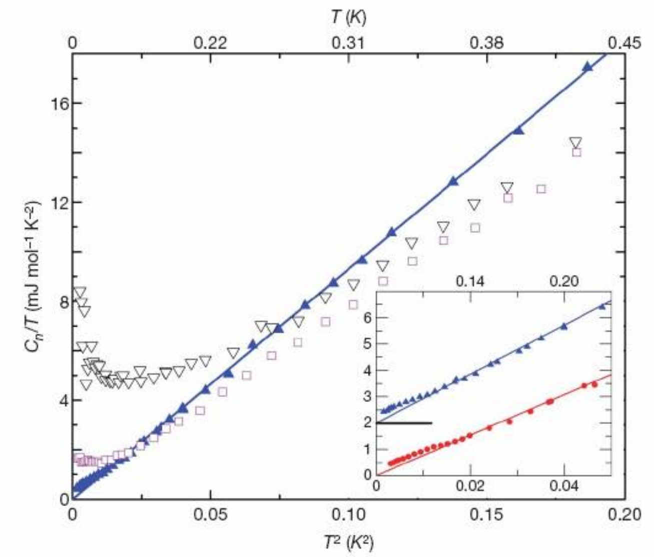

FIG. 5. Plot of C_n/T against T^2. The red circles are for 1 p.p.b. ^{3}He impurity; blue triangles are for 0.3 p.p.m. ^{3}He, purple open squares are for 10 p.p.m. ^{3}He, and black open triangles are for 30 p.p.m. ^{3}He. Inset shows that the 1 p.p.b. and 0.3 p.p.m. (shifted upwards by 2 mJ/mol K) samples both deviate from T^3 curve [15].

plotting the specific heat against T^3 and suggested that besides Debye T^3 term there is a temperature-independent component, as shown in Fig. 6. The data of these four impurities of ^{3}He all fall on straight lines and in particular there are non-zero intercepts for the cases of 10 p.p.m. and 30 p.p.m. samples, which are probably due to the high mobility of ^{3}He impurities.

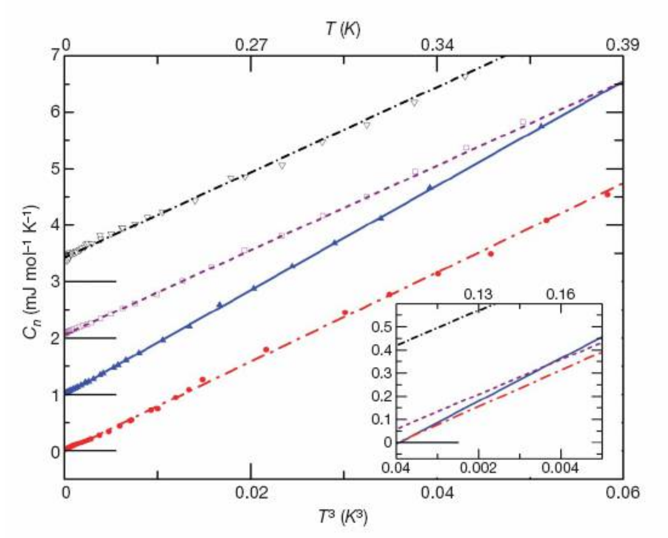

FIG. 6. Plot of specific heat C_n against T^3. The symbols for each impurity concentration are the same as in FIG. 5. Here 0.3 p.p.m., 10 p.p.m. and 30 p.p.m. samples are shifted upwards by 1, 2, and 3 mJ/mol K. Inset shows that 1 p.p.b. and 0.3 p.p.m. samples both have almost zero intercepts but 10 p.p.m. and 30 p.p.m. samples have about 60 and 430 μ J/mol K, respectively, deviation as the constant terms [15].

Figure 7 shows a significant thermodynamic characteristic of the probable second-order phase transition after the subtraction of phonon contribution. For 1 p.p.b., 0.3 p.p.m. and 10 p.p.m. samples broad peaks in heat capacity around 75 mK were observed. Further since the onset of the non-classical rotational inertia for 1 p.p.b. ^{3}He sample was measured to be near 75 mK [13] and the transition temperature in the zero frequency limit might be independent of the concentration of ^{3}He impurities, Chan claimed that the coincidental peaks of non-classical rotational inertia and heat capacity suggested that this is very likely to be a thermodynamic signature of phase transition of superfluid in solid ^{4}He.

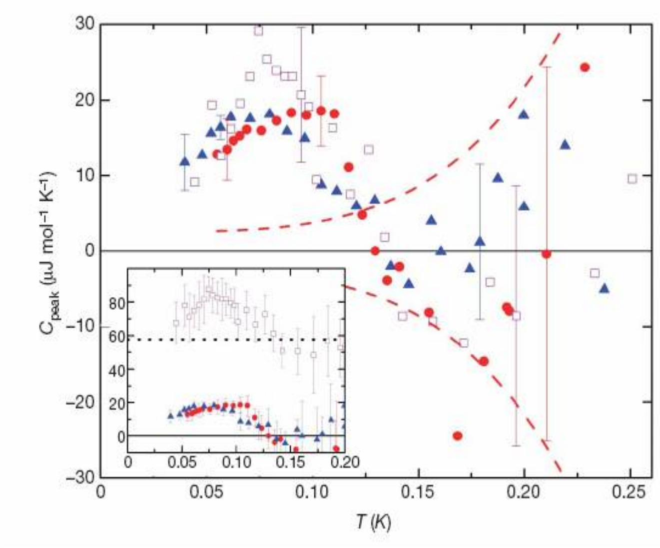

FIG. 7. Specific heat as a function of temperature shows the thermodynamic signature of probable phase transition. The symbols for each impurity concentration are the same as in FIG. 5. When the phonon contribution is deducted, the specific heat presents an impurity-independent peak around 75mK. Inset shows the specific heat for three samples before subtraction of the constant term of 10 p.p.m. sample (For the detail, see [15]).

6 Discussion

With the rapid progress of experimental results, theorists also try to explain these exotic phenomena by using either the microscopic numerical simulation or the phenomenological approaches, for example the Ginzburg-Landau theory and Bose-Hubbard model. Based on the Ginzburg-Landau theory, the ^{4}He phase diagram can be mapped out that the transition from normal solid to supersolid is similar to the experimental measurement by Chan (Fig. 3) and its other phases and phase transitions can also be systematically investigated [17]. When the solid ^{4}He is on the verge of phase transition the free energy describing the coupling interaction between the normal state and superfluid state can be shown as: $f_{\mathrm{int}eracton} \sim \alpha n(x)|\psi(x)|^2$, where the coupling must be repulsive that α is positive, and $n(x)$ is the normal density, $|\psi(x)|^2$ is the superfluid density. If α is small enough the supersolid phase would be stable under low temperature. As a result, at high pressure the solid ^{4}He with quantum zero-point vacancies is incommensurate and the condensation of these vacancies results in the supersolid phase transition. In addition, the phenomenological Ginzburg-Landau theory can also show that the superfluidity is coupled to the elasticity of the crystalline ^{4}He, in which the elasticity would not influence the superfluid transition [18]. Thus in the unstressed crystal there should be a significant thermodynamic characteristic- the λ-transition in heat capacity- in the supersolid transition, as what occurs in the superfluid transition of liquid ^{4}He. This suggestion has been proven by Chan's group (Ref. 15). Another speculative assumption is that the existence of supersolid could be determined by the coupling of strains to the order parameter ψ. If a ^{4}He sample is experienced an anisotropic stress, which affects the coefficient of $|\psi(x)|^2$ in the Landau free energy and is able to make the effective critical temperature positive, this system can perform the supersolid state. This anisotropic stress effect could also be used to account for the experimental results by some groups who fail to find any evidence of the existence of supersolid [19-22].

Another way to explain the experimental results might be using the Bose-Hubbard model. This model has been successfully exploited to study the intriguing behaviors of cold atoms trapped in perfectly optical lattice. However, most condensed matter systems are not so ideal that there exist some imperfections, such as defects or disorders in the crystal. In order to simulate more precisely the experimental system with defects, some modified models are being developed. For instance, the extended Bose-Hubbard model is proposed by adding a spatially extended interaction with Hamilition: $H = -t\sum_{<i,j>}(b_i^+ b_j + H.c.) + \frac{U}{2}\sum_i n_i(n_i - 1) + V\sum_{<i,j>} n_i n_j - \sum_i \mu_i n_i$, where t is the hopping matrix, b_i^+ is the creator of Boson at site i, $n_i = b_i^+ b_i$, and μ_i is the chemical potential. The first term describes the hopping between nearest neighboring atoms and the second one indicates the on-site cost of interaction energy. The third term is used to represent the spatially extended long-range interparticle interactions. By tuning these parameters, the transition between Mott insulator, superfluid, and supersolid states can be systematically studied and the existence of supersolid by the extended Bose-Hubbard model can also be indicated in the mean-field phase diagram [23].

7 Conclusion and Future Work

The probable observation of superfluidity in solid ^{4}He and subsequent experimental studies have rekindled the great interests in this intriguing quantum-mechanical solid. However, even though there are more and more experimental data endorsing the existence of supersolid, the ultimate origin and mechanism are still mysterious and puzzling both experimentalists and theorists. Many speculative theories and arguments have been proposed to explain the supersolidity, but, up to now, none of them are complete and successful to solve the problems either. For the future work, hopefully the nature of this probable supersolid phase transition could be better understood by further investigating the exact shape of λ - transition peak in the heat capacity experiment or by looking for other thermodynamic signatures, for instance the second sound of superfluid. In any case, the experimentalists need theorists to provide more insights and suggestions to explore the future experiment; also theorists need experimentalists' exciting data to come up with a reasonable theory and examine its validity.

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