Mass flux through solid $^4$He induced by chemical potential differences

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Abstract. The thermo-mechanical effect and also direct mass injection have been used to create chemical potential differences between two reservoirs of superfluid $^4$He connected to each other through superfluid-filled Vycor rods in series with solid hcp $^4$He. An increasing DC flux of atoms takes place through the solid-filled cell with decreasing temperature below $\approx 600$ mK. That flux falls abruptly in the vicinity of 75-80 mK, but increases again at lower temperatures. These experiments, done in collaboration with M. Ray, will be briefly reviewed as will our studies of the growth of solid $^4$He where it is seen that it is impossible to add density to a solid freshly made at 60 mK and samples freshly made near 60 mK do not allow mass flux, even when raised in temperature to 200 mK. Solids created above $\approx 300$ mK and cooled to 60 mK accept added density and demonstrate finite mass flux. Relationships to theoretical work and other solid helium work will be briefly discussed.

1. Introduction

A number of years ago it was predicted that solid helium might demonstrate unusual behavior at low temperatures[1], but a search for novel (supersolid) behavior by investigators did not reveal any unexpected behavior[2,3]. These null results diverted attention and it was not until Ho et al.[4] reported unusual behavior in solid helium doped with $^3$He that interest was again heightened in the possibility of a superfluid-like behavior in the solid. Stimulated by the work of Ho et al., Kim and Chan[5] used a torsional oscillator and observed a period shift of the oscillator at temperatures below $\approx 250$ mK. This period shift was interpreted as apparent mass decoupling below a temperature that depended on the $^3$He concentration in the experimental cell. It was suggested that this measurement might signal the presence of a supersolid state. That work[5] was the genesis of the substantial current emphasis on the properties of solid $^4$He.

Since the original measurements of Kim and Chan[5], their basic observation has been replicated in a number of laboratories, but with a large range of inferred values of the superfluid density. The interpretation of the original torsional oscillator experiments continues to be a subject of intense interest and debate. Were a supersolid to be present, it might be expected that such a solid would be capable of supporting a superflow, but early experiments, and more recent ones, in which the solid was directly subjected to pressure gradients (squeezed) failed to reveal any evidence for unexpected flow behavior[2,6].

In our laboratory at the University of Massachusetts Amherst experiments of a conceptually different nature were envisioned. In these experiments, rather than directly squeezing the solid lattice, a sample of solid helium was subjected to an imposed chemical potential difference[7-11]. The result of these experiments was the presence of consistent evidence for a mass flux through a sample cell filled with solid helium below a characteristic temperature. These experiments, done in collaboration
with my colleague Michael Ray, and reported in detail elsewhere [7-11] will be briefly reviewed here along with the presentation of some comments about possible mechanisms that might be responsible for the mass flux that is observed.

2. Experiment – Apparatus and General Approach

The conceptual idea behind the experiments we have carried out is the creation of an environment in which a sample of solid $^4$He can simultaneously be off the melting curve, but still be in direct contact with two separate reservoirs of superfluid $^4$He. Thus, by the imposition of a chemical potential difference between the two reservoirs one would be able to apply a chemical potential difference across the sample of solid helium. This, of course, creates experimental difficulties of various sorts. Our solution to these difficulties is to create a sandwich-like arrangement in which two reservoirs of superfluid $^4$He are in series with two helium-filled porous regions (helium-filled Vycor rods), and the sample of solid $^4$He, as shown in Figure 1. By this means there can be a superfluid-solid interface at pressures above the normal melting curve for $^4$He. A more complete representation of the apparatus is given in Figure 2. With a solid helium sample in the sample cell and appropriate choice of pressure and temperatures of the two reservoirs, R1 and R2, superfluid helium is present in the reservoirs and in the Vycor (V1 and V2).

Experiments of two types can be carried out. (1) In experiments that might be called Type I experiments, with solid $^4$He in the sample cell, if injection of $^4$He atoms to, say, reservoir R1 takes place (with a consequent increase in the pressure of R1, measured as P1), one watches for an increase in the pressure of reservoir R2, P2. If such an increase is observed, it could only have resulted from a flux of atoms through the solid-filled cell[7]. (2) Type II experiments are of a different nature. In these, a temperature difference, T1-T2, is imposed between reservoirs R1 and R2. This results in a fountain effect between the two reservoirs. In this case, one watches for a resulting pressure difference P1-P2, which can only arise if atoms move to restore chemical potential equilibrium in response to the imposed temperature difference[8-10].

In each type of experiment, initially the sample cell is filled with $^4$He (commercial grade, assumed as 300 ppb $^3$He concentration) through capillary 3 (Figure 2). Solid samples can be grown by

![Figure 1. Conceptual diagram of the sandwich-style design of the apparatus. Solid helium is sandwiched between Vycor that contains superfluid helium.](image1)

![Figure 2. Schematic diagram of the apparatus used for these measurements. Reservoirs R1 and R2 are in series with superfluid-filled Vycor and the solid sample.](image2)
means of the blocked capillary technique starting with liquid in region S at a pressure near 55 bar and a temperature near 2.5K. More typically in our work, however, solid samples are grown from the superfluid at a temperature of ~ 350 mK starting at a pressure of 25 bar by means of a simultaneous increase in the pressure above lines 1 and 2 that results in injection of atoms through the Vycor. A complete description of sample growth appears elsewhere[8,11], but later in this brief review we will discuss some unexpected features of such sample growth from the superfluid.

3. Experiment – Type I results.

Type I experiments are experiments in which, with a solid ⁴He sample in the sample cell, the pressure above one line, e.g. line 1, is increased with the addition of atoms, held at that increased value of the pressure (with the source of helium open) for a time, then the source of atoms is closed and the system allowed to come to equilibrium[7]. If the temperature of the solid sample is below ~ 600 mK the pressure measured above line 2 increases and the capacitive gauges on the solid sample (C1, C2, Figure 2) also show increases. Thus, in such a situation helium atoms moved through the solid-filled sample from reservoir R1 to reservoir R2; the sample supports a flux of atoms, with some of the injected atoms retained in the sample thereby increasing its density. For cell temperatures above ~ 600 mK, no flow takes place; the sample is an insulator. Typical examples of these sorts of behavior are shown in Figures 3 and 4; see reference[5]. In experiments of this type we have also studied in some detail the ability of the solid sample to support a mass flux following modest (and sometimes cyclic) changes in the sample cell temperature or pressure, but we will not discuss those results here.

An additional interesting result from these experiments is the fact that in many cases when a sample is grown the two in situ capacitive pressure gauges, C1 and C2, show different values of the pressure. This difference remains constant even in the presence of a finite flux of atoms through the sample cell and increases in C1 and C2. Thus, the solid is able to sustain a pressure gradient. This has implications for precise knowledge of the pressure of a solid sample (certainly so in experiments that involve only one pressure gauge) since the solid is able to sustain differences in pressure in a stable way.

![Figure 3](image1.png)

**Figure 3.** Example of one of the samples that demonstrated a flux of atoms through the solid-filled sample at 358 mK. Note that the growth of pressure P2 in reservoir R2 is nearly linear in time, as might be expected for flux at a critical velocity.

![Figure 4](image2.png)

**Figure 4.** Example of a sample that did not demonstrate a flux of atoms through the solid-filled sample cell. Dwell times as longer than 24 hours showed no flow for temperatures above ~ 600 mK. Additional examples can be found in reference [7].
4. Experiment – Type II results.

With superfluid liquid helium in all areas of the apparatus from R1 through to R2, the two reservoirs are coupled by a series connection through two superleaks, V1 and V2, and a liquid-filled sample cell, S. In such a situation, the imposition of a finite temperature difference between R1 and R2, T1-T2 ≠ 0, is expected to result in a flux of atoms from R2 to R1 to create a mechanical pressure difference and restore chemical potential equilibrium. This is observed with this apparatus in a number of control experiments and the resulting pressure difference P1-P2 ≠ 0 is in quantitative agreement with expectations based on the classic fountain effect in superfluid 4He[8,10].

So, an immediate question is: What will be the case with the sample cell filled with solid helium? That is, does the imposition of a temperature difference result in a flux of atoms to create the appropriate fountain pressure difference, thus restoring equilibrium? The answer depends on temperature in an interesting way[8,10]. To illustrate this, we will first concentrate on the temperature regime above 100 mK where we find that a flux of atoms is present through the solid-filled cell when the temperature is below about ~ 650 mK, consistent with the results found in the case of the Type I experiments. Here a more expansive study of the temperature dependence was carried out. To do so, we considered data like that shown in Figure 5, which is re-plotted in Figure 6a to more clearly show the response of the pressure difference P1-P2 to the imposed temperature difference.

![Figure 5](image_url)  
**Figure 5.** Example of one sample[9] that demonstrated a flux of atoms through the solid-filled sample at 199 mK. The imposition of T1-T2 ≠ 0 results in a flow of atoms and a resulting pressure difference (the fountain effect) to restore equilibrium.

![Figure 6](image_url)  
**Figure 6.** (a) Data as in Figure 5, but here the difference in pressure P1-P2 is shown[7,8]. The slope of P1-P2 vs. time is constant and is found to be temperature dependent and is proportional to the mass flux through the solid-filled cell. (b) Data at a lower temperature (see text)[9,10].

In the example shown in Figure 5 data for the solid sample, designated GT, is shown at 199 mK, first with reservoir temperatures equal at ≈ 1.505 mK, and then with a sequential change in the reservoir temperatures, first by an increase in T1, followed by a return to T1 = T2, and then an increase and subsequent decrease in T2. In response, the pressures in reservoirs R1 and R2 change as a result of the imposed chemical potential difference and a flow of atoms through the solid-filled cell results to restore chemical potential equilibrium. The resulting pressure difference, P1-P2, Figure 6a, is linear in time, which is consistent with a flux of atoms through the cell that is limited by a temperature-dependent critical flux and independent of the applied chemical potential difference imposed by T1-T2 ≠ 0. Of course any bottleneck could limit the flux and we have determined by
studies of flow through Vycor that the Vycor does not impose any bottleneck, except near 100 mK. Further details can be found in [10].

The temperature dependence of this flux above 100 mK is shown for a solid sample[9,10] at 26.03 bar in Figure 7. These data can be reasonably well fit by an expression of the general form flux \( \sim \exp(-T/a) \). We will return to discuss this temperature dependence later in this review. Cooling the apparatus below 100 mK results in a rather dramatic and unexpected behavior (Figure 8)[9,10]. In the vicinity of 75 mK the flux decreases by more than an order of magnitude over a very narrow temperature range (\( \sim 5 \) mK) and then rises again at lower temperature (Figures 6b, 8). This behavior was observed with several samples, but not with all samples. So, the presence of this behavior apparently depends on the particular sample under study. And, as described in detail in [10], while the flux is stable during measurements at most temperatures, in the vicinity of this strong flux minimum the flux is not stable and changes as time evolves. So, there is some apparent metastability or dynamic behavior present in this narrow temperature range near 70 mK. And, it may be that the history of the sample influences the value of the flux [10]. At this writing it is not clear what the presence of unstable behavior means, but it may point to a region in which there is a transition from one mechanism by which the flux is carried by the solid to another one that is present at lower temperatures. A substantial discussion of the fluctuation behavior of the flux in this temperature regime is presented in reference [10].

![Figure 7](image1.png)

**Figure 7.** A measure of the flux through the solid-filled sample cell as a function of temperature determined from data similar to that shown in Figure 6 for a sample at 26.03 bar.

![Figure 8](image2.png)

**Figure 8.** The flux measured as a result of cooling the sample cell below 100 mK. More comprehensive results are presented in reference [10].

5. Experiment – The Growth of Solids

During our experiments it was typical to grow our solids from the superfluid by adding mass through the Vycor rods. In a typical such solid growth, the temperature would be \( \sim 350 \) mK and from a pressure just below the melting curve the pressure would be increased simultaneously by opening reservoirs R1 and R2 to a source of high-purity \(^4\)He gas. By this method it was possible to grow solid samples to pressures within the range we were able to investigate, but we could not cause such growth at all temperatures. For example, we found that if we attempted to grow a solid sample by this method at 60 mK it was not possible and the sample would not grow off the melting curve, even for an over-pressure of more than one bar. For such a sample, an increase in the temperature to 100, or even 200 mK also did not allow the sample to grow; dwell time as long as \( \approx 24 \) hours resulted in no growth.
And, for such a sample on the melting curve, it was not possible to observe any flux. But, if the temperature of this sample was raised above \( \approx 300 \) mK, the sample could readily be grown. And, once grown, it could be cooled and at lower temperature grown further. Once grown, such a sample always showed a finite flux\( [9,10] \).

One measure of the ability to grow a sample above the melting curve is the shift in the capacitance, \( \Delta C \), that results from a given step in the applied pressure, \( \Delta P \). The quantity \( \Delta C / \Delta P \) can be interpreted as a measure of what has been called the isochoric compressibility and some measurements of this quantity are shown in Figure 9\([11,12]\). Also shown on the figure (in red) are some cases for which it was not possible to initially grow the solid above the melting curve.

Another interesting feature seen during solid growth is transient pressure decreases documented by the in-situ capacitive pressure gauges, which are accompanied by transient increases in the temperature of the sample cell. As example of this behavior seen during growth at 260 mK is presented in Figure 10. A study of the details of behavior of this sort results in the conclusion that these transient events result from the solidification of small liquid regions in the solid\([11]\).

6. Discussion

A theoretical understanding of these phenomena requires there to be an understanding of several types of behavior: (1) the failure of growth from the superfluid when such growth is initiated below about 250 mK; the ability to grow solid from the superfluid if the sample cell is above this temperature, (2) the presence of a mass flux through a solid-filled cell for temperatures below about 650 mK, (3) the increase in the density of the solid that is always present when a finite mass flux through the solid-filled cell is observed, (4) the dramatic temperature dependence (flux minimum) seen in the vicinity of 75 mK, (5) the transient behavior seen in the vicinity of the flux minimum and (6) the increase of the flux with decreasing temperature for temperatures below about 70 mK. Also relevant, of course, is a need for an understanding of the relationship among these results and those many results obtained by torsional oscillator, shear modulus and other techniques.

It seems clear from recent theoretical work that the mechanism that might lead to supersolidity originally proposed is not a viable mechanism\([13-15]\). A number of scenarios have been
suggested. One possibility that appears consistent with a number of the above observations is the possibility of the superclimb of edge dislocations in the solid and the presence of a super-flux along the dislocation cores[16]. This suggestion is also consistent with the growth in density of the solid in a temperature window and the presence of a super-flux for temperatures below a characteristic temperature. As discussed in reference [10], if we assume edge dislocations carry the flux, then we can write the mass flux as \( \frac{dm}{dt} = \xi v \rho N A \), where \( \xi \) is the superfluid density, \( v \) is the flow velocity, \( \rho \) is the density (assumed to be equal to the density of the solid), \( N \) is the number of dislocation edges that conduct and \( A \) is the effective cross sectional area of one dislocation. For a typical mass flux of \( 2.9 \times 10^{-8} \) g/sec, with \( A = 1 \) nm\(^2\) and \( \xi = 1 \), we find that \( vN = 1.7 \times 10^7 \) cm/sec. If the velocity is limited by the velocity of sound then \( v \approx 350 \) m/sec, but if it is limited by the velocity found to be characteristic of the torsional oscillator experiments, then \( v \approx 10 \) \( \mu \)m/sec. In these two extremes we find \( N = 485 \) or \( 1.7 \times 10^{10} \), which correspond to a dislocation density of \( 1600 \) cm\(^{-2}\) or \( 5.6 \times 10^{10} \) cm\(^{-2}\) at the other extreme.

A strong valley in the flux at a characteristic temperature is a feature of non-linear behavior found theoretically[17] for edge dislocations, but to date it has not been possible to conclusively test this scenario due to limitations in the existing apparatus that prevent precise measurements for much smaller values of the imposed chemical potential difference. That is, if this mechanism is present the current experiments are in the regime of strong non-linearity.

One can speculate about a number of intriguing possibilities. For example, there may be two entirely different mechanisms at work in the solid, with a transition in behavior between the two mechanisms near 75 mK. So, for example, perhaps it is the case that dislocations provide the super-flux pathways through the solid. In this case we speculate that it might be possible that as the temperature is lowered below \( \sim 80 \) mK the pinning of \( ^3\)He to the dislocations could reduce the flux along them as the temperature is lowered by inhibiting the ability of the flux cores to support the flux. If at even lower temperatures there were to be a rearrangement of the dislocation network, this might cause the flux to increase again due to the creation of detours around the \( ^3\)He-induced bottlenecks with the result that there is an unusual temperature dependence to the measured flux[10]. This is clearly speculative since it is not clear that such a rearrangement would take place.

Continuing to speculate, it is interesting to ask what other possibilities might explain the flux behavior in the higher temperature regime (Figure 7) where the temperature dependence can be reasonably well fit to an expression of the form flux \( \sim \exp(-T/a) \). This is an intriguing and unusual temperature dependence. If one carries out fits to a functional dependence of that form applied to data like that shown in figure 7[9,10], one finds that \( a \approx 160 \pm 20 \) mK for data at 26.0 bar. Although the range of flux values that can be fit is relatively modest \( \sim 25:1 \), the fits to the data are reasonable. Recent simulations of interacting bosons in one dimension indicate that conditions may exist in which the superfluid density can have precisely this form[18] and screw dislocations have been shown to have Luttinger liquid-like behavior[19]. But, such a scenario would seem to require rather short lengths of whatever is carrying the flux and require many short-segment interconnections to carry a flux across the macroscopic sample. Perhaps the solid demonstrates Luttinger liquid behavior at higher temperatures and then makes a transition to different behavior below about 75 mK, with only the lower temperature behavior visible to torsional oscillator experiments.

### 7. Future work

A number of possible future experiments of this general sort come readily to mind. For example, it seems important to carry out similar experiments with a relatively pure \(^4\)He sample as well as with a number of samples with higher \(^3\)He concentration. We also wish to modify our apparatus to allow measurements to lower temperatures that we have been able to achieve to date. And, to test the notion
that non-linear behavior is responsible for the strong valley in the temperature dependence of the flux near 75 mK we would like to be able to precisely control and measure the response to very small applied chemical potential differences. This would also allow us to explore more details about the presence of a critical flux. Finally, we have been limited by our technique to a relatively small range of pressures and we would like to expand that range by variations of the basic technique that we have used to date. This will require us to substitute other porous material for the Vycor or utilize a somewhat different experimental technique.

8. Conclusion

In conclusion, we have seen clear evidence for mass flux through solid $^4$He in response to the application of a chemical potential difference using two approaches. This flux has the behavior of a critical flux and has substantial temperature dependence. In particular, there may be two regimes of behavior. The origin of this behavior is not yet clear, but the experiments support the possibility that superfluid-like behavior permeates the solid. Connection to the torsional oscillator and shear modulus[20], solid stiffening[21] and rotation[22] experiments is also not yet clear, but it is possible that the lower temperature behavior we observe has a similar fundamental origin. Indeed, there are alternate explanations[23] for the unusual phenomena seen in solid helium and the field is very active, dynamic and controversial at the moment with a number of other interpretations[24-26].

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