SUPERFLUIDITY IN ^{3}HE: DISCOVERY AND UNDERSTANDING

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by

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1. DISCOVERY

In starting to compose this lecture, I am reminded of the general excitement which permeated the field of low temperature physics in 1971. There were new cooling technologies being developed, and everyone felt that interesting and important new physics was just waiting to be discovered in the ultra-low temperature world to be made available for study by these techniques.

I had come to Cornell to do graduate work in what was then called solid state physics, but was quickly attracted to the promising field of low temperature physics by a talk which Bob Richardson gave in the fall of 1967, in which he described how dilution refrigerators work. At the time, I was working as a teaching assistant for Dave Lee. Dave seemed to think I was fairly bright, and invited me to join the group. By the end of my first year at Cornell, Jim Sites, a senior graduate student, and I had built the dilution refrigerator which would later be used in the discovery of superfluidity in ^{3}He.

A dilution refrigerator is a device which utilizes the non-vanishing (~6%) solubility of liquid ^{3}He in superfluid ^{4}He at low temperatures to effectively 'evaporate' liquid ^{3}He down to arbitrarily low temperatures. Because ^{3}He at mK temperatures is a degenerate Fermi fluid whose entropy is proportional to the temperature, such a device has a cooling capacity which decreases at least as fast as the square of the temperature. In those days, such refrigerators could only reach about 15 mK.

Sites intended to measure the magnetic susceptibility of solid ^{3}He down to as close to the nuclear spin ordering temperature as possible, expected to be at about 2 mK. The effective spin interactions in this system result from atom-atom exchange at rates as high as 40 MHz. By contrast, one calculates an atom-atom exchange rate for silicon which is less than one over the age of the universe! The dilution refrigerator was only the first step in Jim's new cooling process. The second refrigeration stage was to be through the adiabatic solidification of liquid ^{3}He, or Pomeranchuk refrigeration, named for the Russian theorist that had suggested the process1) in 1950, before anyone had even created liquid ^{3}He! Pomeranchuk reasoned that liquid ^{3}He would become a degenerate Fermi fluid at low temperatures owing to its half-integral spin. Thus at sufficiently low temperatures its entropy would depend linearly on temperature. The solid would possess an entropy dominated at low temperatures by the disordered nuclear spins, S_{solid}~Rln2, and hence at sufficiently low temperatures the liquid entropy would drop below that of the solid. In this rather unique situation, the latent heat of melting would be negative, and hence if one compressed to solidify part of a liquid sample at constant entropy, the liquid would have to cool. The actual cooling rate is about 1 mK for every percent of the liquid sample converted to solid. Pomeranchuk had argued that this process would continue down to the temperature at which the solid spin system ordered, making it an ideal vehicle for studying nuclear ordering in the solid.2)

The main problem with Pomeranchuk's proposal was that the work done on the system to solidify the liquid, $P_{melt}\bullet(V_{solid}-V_{liquid})$, exceeded the latent heat by two to three orders of magnitude near the solid ordering temperature. How reversible would the compression process be? Anufriyev in Russia had showed that the process worked3) in 1964, but he only reached a temperature of about 20 mK in his experiments. No one had demonstrated that this process could reach temperatures significantly below those available with a dilution refrigerator, much less down to the solid ordering temperature. Both John Wheatley's group at La Jolla (U.C. San Diego) and we at Cornell were betting on positive results. Despite these and other complications, Sites' experiment worked4) during my second year of graduate study, and Jim graduated in the summer of 1969, leaving his cryostat to me. Jim was the only member of the low temperature group to obtain a Ph.D. in just four years.

In my third year, I set about to improve the dilution refrigerator Sites and I had built, and also built a new Pomeranchuk cell based on a design I had developed the previous year, while recovering from knee surgery following a skiing accident. Sites had used a fairly complicated arrangement of three helium filled chambers and two metal bellows to decrease the volume of the ^{3}He region, necessary to compress and solidify the liquid. In his cell the convolutions of the bellows containing the ^{3}He closed as compression occurred. If solid formed in the convolutions, plastic deformation of that solid, and hence irreversible heating, would occur. My cell was simpler, relying on only two helium chambers with bellows in the form of a hydraulic press as shown in Figure 1. Superfluid ^{4}He was introduced under pressure to the large bellows assembly at the top, forcing the central piston downward, and extending the lower bellows into the ^{3}He filled region. A photograph of the upper bellows attached to the dilution refrigerator is shown in Figure 2.

Beginning my fourth year of graduate study, the new double refrigeration stage was operational, and I had also improved our NMR thermometry scheme. To utilize the system to study physics, something I was not yet very comfortable doing on my own, I teamed up with Linton Corruccini, a student in the group who was one year ahead of me. Linton wanted to test an unusual prediction by Leggett and Rice5) resulting from the effects of molecular fields in the collisionless regime arising from Fermi liquid behavior of ^{3}He, in which the spin diffusion coefficient in the liquid would vary with the angle by

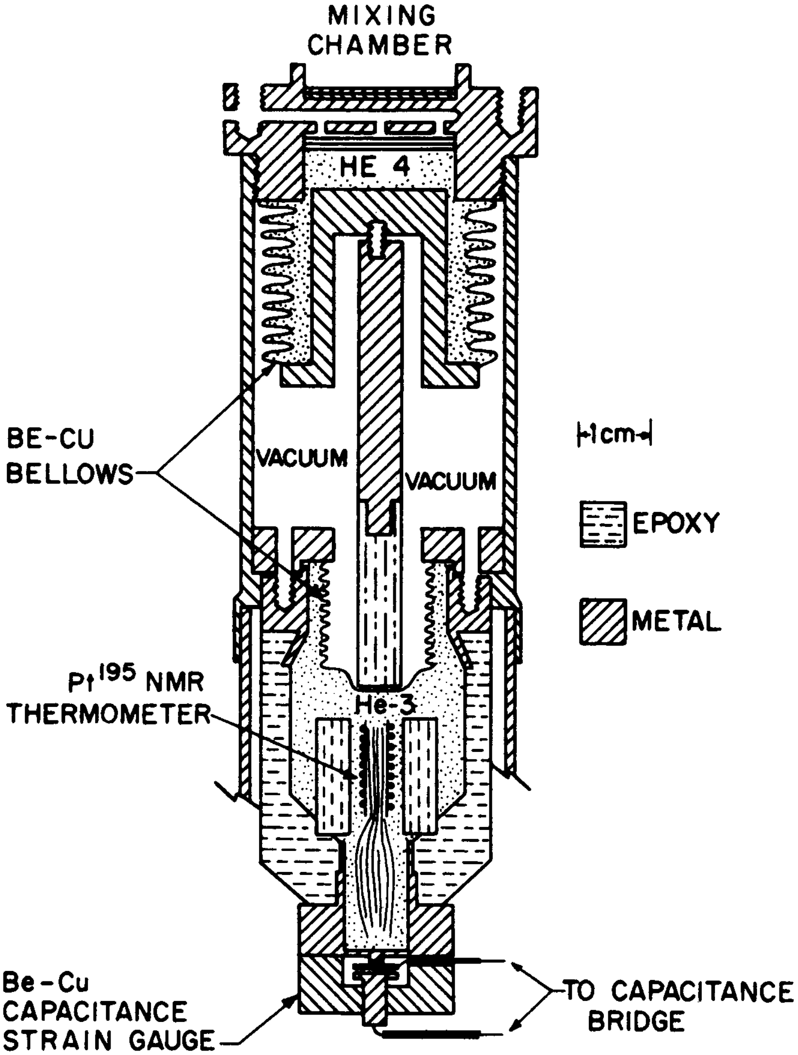


Figure 1: Schematic diagram of the Pomeranchuk cell used in the discovery of superfluidity in ^{3}He. Superfluid ^{4}He is added to the upper bellows assembly under pressure, forcing the lower bellows into the ^{3}He-filled region. The capacitive pressure transducer is at the bottom. (Reproduced with permission from reference 11).

which the spins where tipped in a pulsed NMR experiment. We used the Pomeranchuk cell to indirectly cool a separate sample of liquid ^{3}He down to about 6 mK. The experiment worked$^{6)}$, and we were able to estimate the Landau Fermi liquid parameter $F_1{}^a$ for the first time using the Leggett-Rice theory.

It was soon to be my fifth and presumably final year of graduate study. I had married Phyllis S. K. Liu in August of 1970, just two weeks after she successfully defended her Ph.D. thesis, and she was working as a postdoc in biochemistry waiting for me to complete my thesis. Dave Lee had wanted me to detect nuclear spin ordering in solid ^{3}He using my apparatus. I wasn't exactly sure how to do this, but before long Dave showed me a preprint of an article

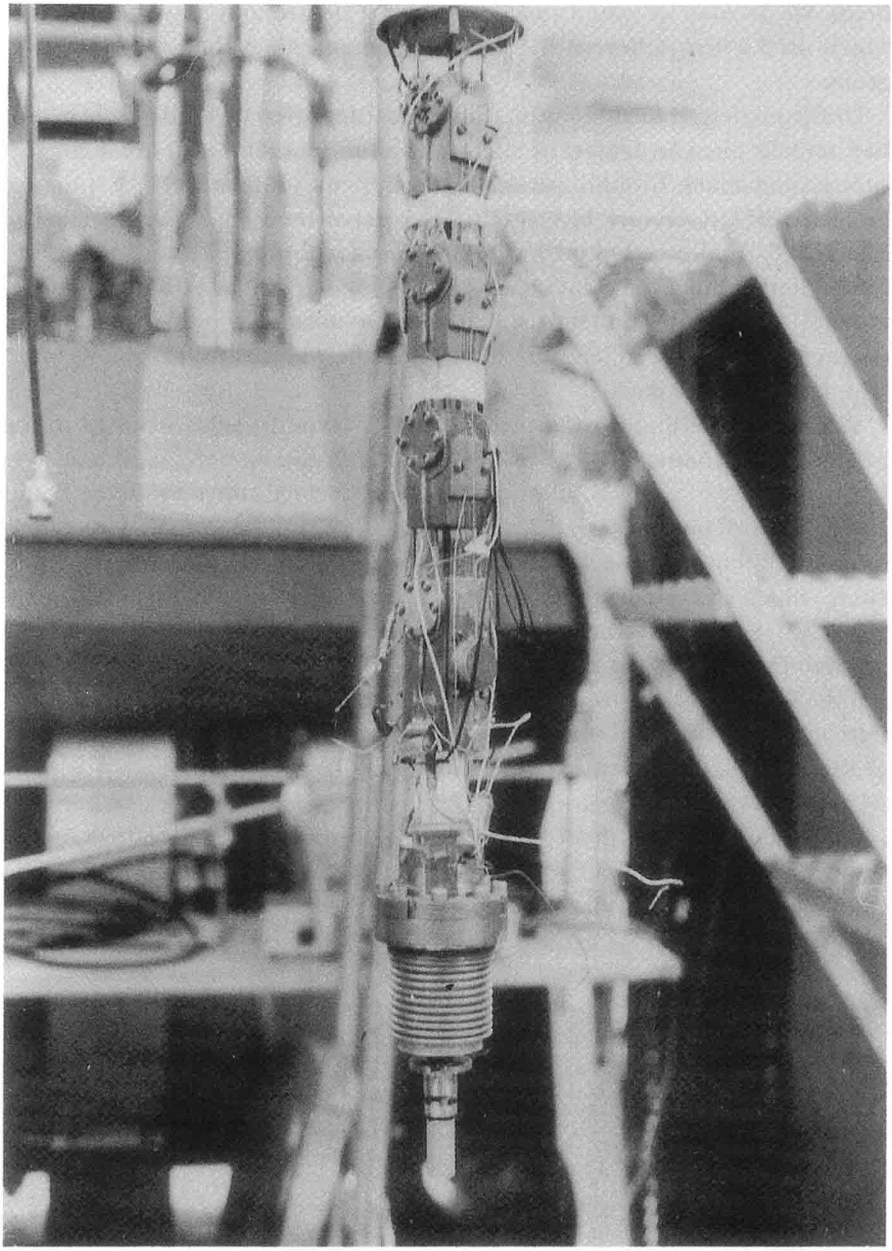


Figure 2: Photograph of the lower portion of the dilution refrigerator used in the discovery, showing the heat exchange column and, at the bottom, the mixing chamber with the ^{4}He bellows chamber and piston for the Pomeranchuk cell attached.

by John Wheatley's group(7) at U.C. San Diego in which the depression of the ^{3}He melting pressure was measured as a function of temperature at several magnetic fields. This depression should be a direct measure of the difference in magnetization between the Fermi liquid and the solid. The group's results suggested an anomalously large magnetization in the solid at low magnetic fields. We decided we would check this unexpected result with our apparatus, which used a very different thermometry scheme than that of the La Jolla group.

Unfortunately, it turned out that the effect reported by the La Jolla group had actually been an artifact of that group's thermometry, and the degree of suppression which I found was small and difficult to measure8). Despite the fact that this did not look like a good thesis experiment, I persisted doggedly. Finally Bill Tomlinson, a postdoc in the group, and Jim Kelly, another graduate student, argued that I had monopolized the lab's only NMR magnet long enough, and that it was their turn to use it. I reluctantly agreed to give up the magnet, but kept my apparatus cold in case their apparatus leaked, as often happened in those days.

While I was waiting for the verdict on their experiment, I decided to see just how low a temperature I could reach with my Pomeranchuk cell. We knew our copper wire NMR thermometer lost thermal contact with the liquid ^{3}He in the cell below about 2.7 mK, but I felt we could extrapolate our thermometry to lower temperatures using the expected slope of the ^{3}He melting curve, which had already been measured at La Jolla9) and by myself to below 3 mK. My experiment consisted of forming solid ^{3}He at a very steady rate, and plotting the melting pressure vs. time on a strip-chart recorder. It is important to note here that I was using a capacitive pressure transducer of the sort first developed by Straty and Adams10) at the University of Florida. In such a device the hydrostatic pressure flexes a thin metal diaphragm to which one plate of a parallel plate capacitor is attached, thus changing the capacitor gap, which was measured with an AC capacitance bridge. It had far better resolution than anything which had been available before. The parts of my pressure transducer are shown in Figure 3.

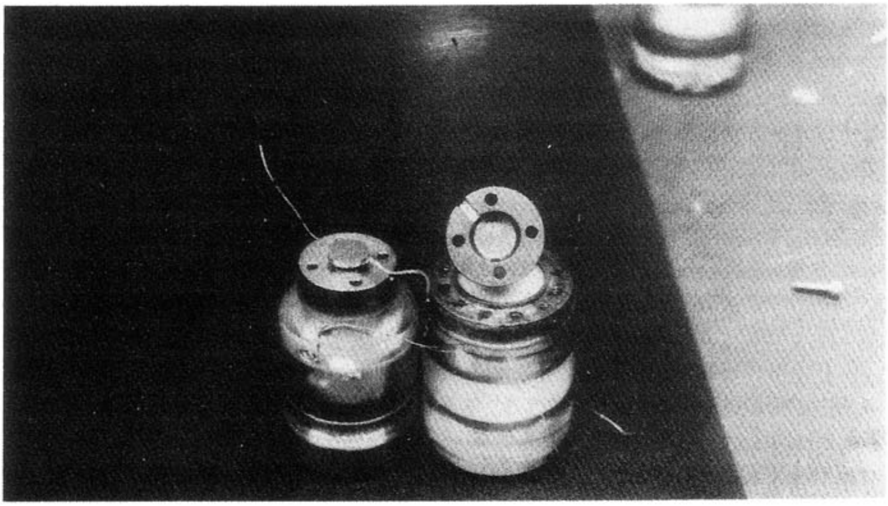


Figure 3: Photograph of the capacitive pressure transducer for the ^{3}He cell during assembly. The moving capacitor plate attached to the metal diaphragm is seen at the left, while the stationary plate is on the right.

The first such experiment was carried out on November 24, 1971, the day before the American Thanksgiving holiday. As I watched, the pressure rose steadily as the cell cooled. Suddenly, at a temperature I estimated to be about 2.6 mK, the rate of cooling abruptly dropped by about a factor of two. I guessed that this decrease in the cooling rate signaled the onset of heating due to the plastic deformation of solid ^{3}He by the moving bellows, and soon decided to terminate the compression. A portion of the resulting pressurization curve first showing this 'kink' is seen in Figure 4. The handwritten numbers in the figure were added four days later. After melting the solid in the cell by decompression, I decided to let the cell pre-cool to as low a temperature as could be reached with my dilution refrigerator over the entire four-day holiday, and then to try the experiment again on Monday. If I started at 15 mK rather than 20 mK, there would be 30% less solid in my cell at 2.6 mK than there had been in the compression on Nov. 24.

On that fateful Monday I got into the lab at about noon, ate a quick lunch as was my habit, and started the compression at about 12:35 pm. By 5:50 pm I neared the pressure at which the sudden decrease in cooling rate had been

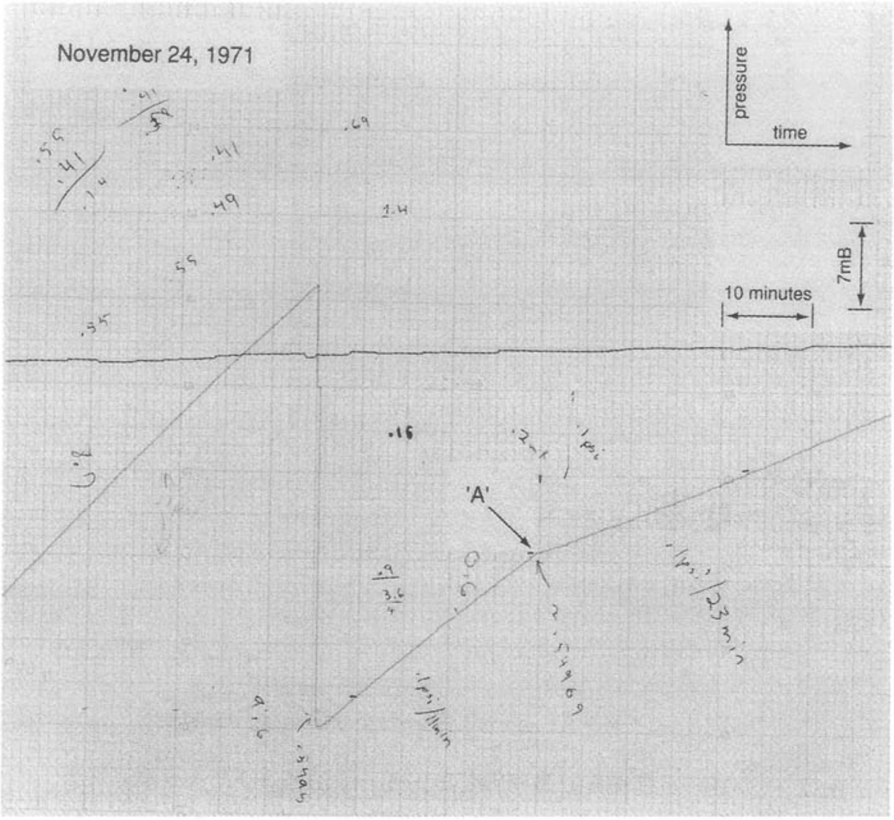


Figure 4: Pressurization curve taken Nov. 24, 1971, showing the first observation of the 'A' transition. Pressure increases vertically while time increases to the right. The abrupt jumps in the pressurization curve occurred when the capacitance bridge was re-balanced. The ragged line is the temperature of the dilution refrigerator.

seen in the previous run. I did not expect the kink to occur at the same pressure, if at all. Nonetheless, I soon saw another kink in the pressurization curve, and could tell that it was close to the same pressure at which it had occurred before. My heart sank. I then made a careful determination of the pressures at which these 'glitches' had occurred, and found the two pressures were the same to within about one part in 50,000!

At this moment adrenaline began to flow through my veins, as I immediately recognized that the probability that plastic deformation would just begin in my cell at exactly the same pressure on successive compressions with very different starting conditions was vanishingly small. A more logical explanation for this coincidence was that this glitch signaled some highly reproducible phase transition in my cell. Had I managed to reach the temperature of the nuclear magnetic phase transition in solid ^{3}He? The temperature seemed too high. I then repeatedly compressed and de-compressed through the region of the glitch to insure that it was indeed repeatable, and to measure its pressure more accurately. The initial pressurization curve through the 'glitch' that day is shown in Figure 5. I then found Bob Richardson, and we discussed the possible nature of the new transition I had discovered. We agreed that if there was a first order transition in the solid in which the spin system lost per-

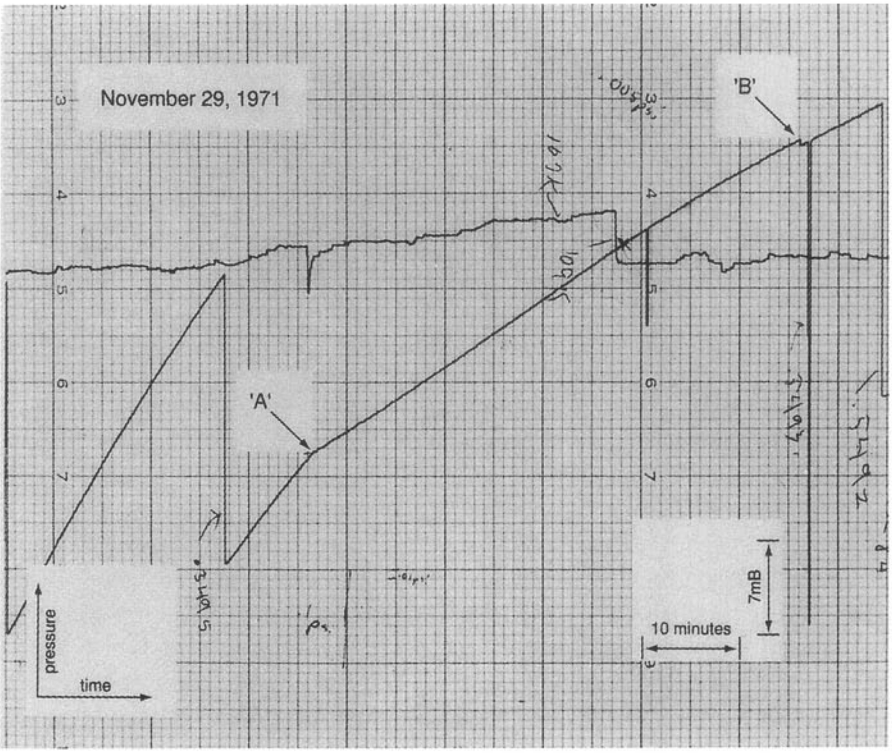
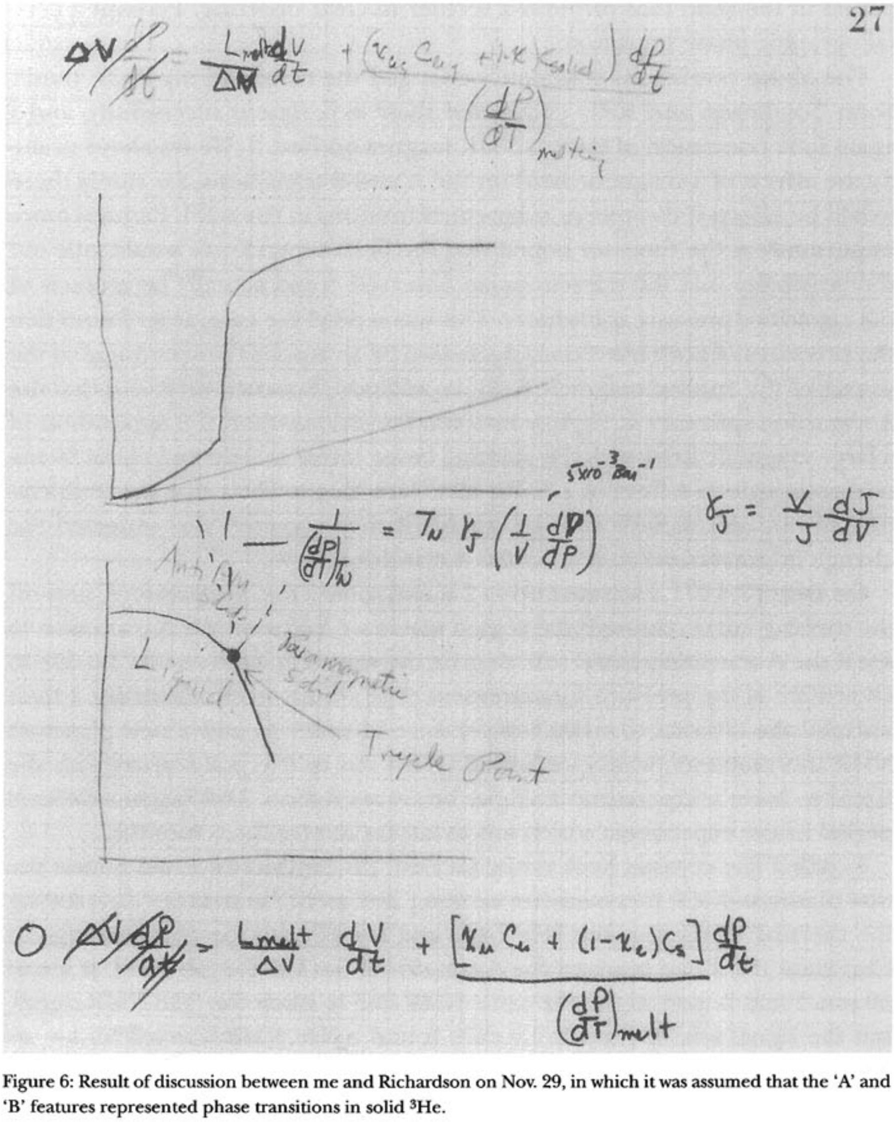


Figure 5: Pressurization curve taken Nov. 29, 1971, showing the second observation of the 'A' transition. Note that the 'A' features in this and Fig. 4 occur at the same pressure, and note the tiny but abrupt drop in pressure labeled 'B'. This was the first time the 'B' transition was ever observed.

haps 30% of its entropy, we could understand the change in slope of the pressurization curve. This discussion resulted in a possible magnetic phase diagram for the solid spin system being recorded in my lab book, which is shown in Figure 6. There was no discussion what-so-ever at that time of a possible phase transition in the liquid!

Note that at the far right in Figure 5 one can see a small abrupt drop in the pressure vs. time curve. I soon realized that this feature, which was always seen, but not at the same pressure, was also a transition. But this transition exhibited a substantial degree of supercooling. There was always associated with this tiny back-step a narrow plateau in the de-pressurization curve upon warming, and this plateau did occur at a highly reproducible pressure. This apparent transi-



$$\cancel{\Delta V}\cancel{\frac{dP}{dt}} = \frac{L_{\text{melt}}}{\Delta M}\frac{dV}{dt} + \frac{\left(x_{liq}C_{liq} + (1-x)C_{solid}\right)\frac{dP}{dt}}{\left(\frac{dP}{dT}\right)_{\text{melting}}}$$

$$\frac{1}{\left(\frac{dP}{dT}\right)_{T_N}} = T_N\,\gamma_J\left(\frac{1}{V}\frac{dV}{dP}\right) \qquad \gamma_J = \frac{V}{J}\frac{dJ}{dV}$$

$$\cancel{\Delta V}\cancel{\frac{dP}{dt}} = \frac{L_{\text{melt}}}{\Delta V}\frac{dV}{dt} + \frac{\left[x_{l}C_{l} + (1-x_{l})C_{s}\right]\frac{dP}{dt}}{\left(\frac{dP}{dT}\right)_{\text{melt}}}$$

Figure 6: Result of discussion between me and Richardson on Nov. 29, in which it was assumed that the 'A' and 'B' features represented phase transitions in solid ^{3}He.

tion seemed far more difficult to explain by our model of the solid spin system. While we had originally called the higher temperature transition a 'glitch' and this lower temperature transition the 'glitch prime', we soon renamed the 'glitch' the A transition, and the 'glitch prime' the B transition. Those designations remain to this day, and as we shall see, they were actually quite prophetic of the microscopic identities of the two superfluid phases.

We soon found that if we continued compressing after the A and B transitions, the pressure would continue to rise to a maximum value, though slowly, and that the total change in pressure from the pressure of the A transition exceeded the change possible if the A transition was the 'expected' solid spin ordering transition, or even if it was only the 'expected' transition at 2 mK. The solid seemed to hold onto its entropy to too low a temperature. To kill two birds with one stone, we assumed that at the A transition something happened in the solid that prevented further nuclear ordering. Perhaps a crystallographic phase transition.

The above revelations completely changed the course of my Ph.D. thesis. Soon Tomlinson and Kelly completed their experiment successfully, and I again took possession of the lab NMR magnet on Dec. 2. We set about to study the effects of a magnetic field on the A and B transitions, for surely there would be effects if these were magnetic transitions in the solid. Perhaps more importantly at the time, we hoped that the field dependence would rule out the possibility that the transitions we observed could actually be artifacts of the capacitive pressure transducer. This was indeed the case, as we found that the pressures of both transitions decreased by an amount proportional to the square of the applied magnetic field. In addition, however, we found that the A transition split ever so slightly into two transitions upon the application of a large magnetic field, with the splitting being linear in field and about 60 microkelvin wide in a field of 1 T. We also were able to show that the temperature in our cell, as indicated by our NMR thermometer, also reflected the change in pressurization seen at the A transition.

On Dec. 17, 1971, I warmed up to 1 K and moved the ^{3}He pressure, now off the melting curve, through the region where we had seen the A transition to see if the A transition could still, despite the magnetic field results, be due to an artifact of the pressure measurement. The results were reassuring. I then warmed the cryostat to room temperature in order to add a new platinum NMR thermometer, which we hoped would stay in thermal contact with the liquid to lower temperatures and give better resolution. This began a difficult period in the experiment which was to last for almost three months.

I cooled the cryostat back down on Dec. 21. By Dec. 24 I had gotten the new platinum NMR thermometer working and spent the next few days testing for thermal contact to the ^{3}He bath and for rf eddy current heating. By Christmas Eve I had reached the A transition, but left the lab early, at about 10 pm. I had hoped to use the same NMR coil to study the ^{3}He NMR signal, but the signal was very small. I finally found a ^{3}He NMR line a 1:55 am on New Years night. It seemed pretty useless. I left the lab at about 3:00 am, and when I came in the next afternoon someone had written "Happy New Year, Doug" in the lab book. Two days later we compressed and found that very little solid ^{3}He formed between the platinum wires of the NMR thermometer. We could not use a single NMR coil to measure temperature and simultaneously study the ^{3}He NMR signal!

On January 5 I flew to New Jersey to interview for a job at Bell Laboratories, fully believing that the A and B transitions were in the solid nuclear spin system. Luckily, no one argued those identifications, and within two months I was offered a permanent job as a regular member of their technical staff. At this point we decided to write up our results, without the NMR confirmation of our interpretation, and submitted to Physical Review Letters a paper entitled "Evidence for a New Phase of Solid ^{3}He". This paper sailed through the peer review process and was quickly published$^{11)}$.

On Jan. 21 I warmed up again to replace the ^{3}He cell with one which had a separate ^{3}He NMR coil. We had tried a new epoxy resin for this cell, and it cracked as we cooled down. I then made another cell with the more traditional 'Stycast 1266' resin, and replaced our beryllium copper pressure transducer with one made of 304 stainless steel, to further ensure that the effects we had seen were not simply an artifact of our strain gauge.

I cooled down again on Feb. 10, but started having problems with the dilution refrigerator. Later it became clear that corrosion due to the solder flux we used had nearly occluded the fine capillary tubes which connected the heat exchangers together. I also had troubles with the NMR electronics. Finally, on Feb. 18 (my wife's birthday) at 11:58 pm I wrote in the lab book: "I've got the ^{3}He NMR line–it's a big mother!!" (I didn't think back in those days that these volumes might have historical significance!) This euphoria was short lived, however, as two days later, Cornell University lost electrical power, allegedly due to a squirrel shorting out a high voltage power line, and the tube which pumped on our ^{4}He pot became blocked with solid air. I had to warm up to 77 K to eliminate the block, but when I again transferred liquid helium into the dewar, the cryostat had sprung a leak in the main vacuum flange which admitted the pumping lines to the experimental vacuum space. The leak defied my every effort to eliminate it, and in late February I began to machine the parts necessary to replace the main vacuum flange, a very difficult and risky procedure.

At this point Willie Gully, a second year student whom Dave Lee had assigned to work with me, asked if he could try his hand at fixing the leak. Up to this point I had made every effort to insure that Willie's 'uneducated' hands stayed off my precious cryostat. (This unfortunate attitude persists amongst my own senior graduate students to this day.) Since I had already written off the vacuum flange, however, I told Willie to go ahead. I had already tried the old Wheatley trick of applying a warm mixture of glycerin and soap (Ivory Flakes) to the offending joint, but Willie used a different mixture, and poured a rather large beaker of warm glycerin and soap over the entire vacuum flange, to a depth of about 1 cm as I recall. When we cooled back down on Feb. 29, the leak had vanished, never to return. From that day forward, Willie was a full collaborator in the project.

I took NMR data throughout the first two weeks in March, recording the NMR peak height as a function of time as we cooled and warmed through the A transition. In the ^{3}He cell the liquid contribution to the NMR signal was small, and expected to be independent of temperature, while the solid signal rose as one cooled almost as 1/T. The stainless steel pressure transducer produced a fairly severe magnet field gradient, but we were clearly able to show that the growth rate of the ^{3}He NMR peak, presumably due to the increasing solid signal, increased slightly just as we cooled through the A transition; every time. The change was not large, but consistently present and highly correlated with the kink in the pressurization curve. Much later we were to realize that this increase in growth rate occurred because the liquid NMR frequency shifted as we cooled through the A transition, and superimposed itself on top of the solid NMR signal.

By this time we were becoming increasingly aware of the possibility that the A transition might be in the liquid, and I considered the possibility that a change in the thermal conductivity of the liquid at the A transition could result in a change in the rate of solid growth in the region seen by the NMR coil, thus producing the small change in peak growth rate discussed above. To rule out this possibility, we decided we needed to have spatial resolution, and I began working on a fourth cell design. In this new cell, the ^{3}He NMR coil consisted of five separate solenoidal coils, each oriented vertically, over a total length of about 2.5 cm. The idea was to look at solid formed in each of the five coils. To differentiate the signals from the various coils, I put soft iron shims between the pole faces of the NMR magnet, so that the (horizontal) NMR field would be larger at the bottom of the cell than at the top. Then the NMR signal from each coil would occur at a different frequency. We cooled this cell down on March 27, and I saw the expected five NMR peaks on April 3, but warmed up once more, to install a much better, final modification to the ^{3}He cell. In reading the lab books 25 years later, I am amazed at how often I was willing to thermally cycle such a fragile piece of apparatus!

I had by this time spoken with Michael Fisher about how one would expect solid ^{3}He to grow in a Pomeranchuk cell. Almost everyone imagined solid nucleating spontaneously at the warmest point in the cell (since there the pressure would be above the melting pressure), resulting in ^{3}He snow. Michael gently told me about the surface energy which would exist at the solid-liquid interface, and how it would act to prevent spontaneous solid nucleation. He expected the solid to nucleate only on surfaces, and at only a few places in the cell, with all subsequent solid growing from these seeds. To take advantage of this growth characteristic, I once again changed the ^{3}He NMR coil, this time to a single vertical solenoid roughly 0.5 cm in diameter and 2.5 cm long. In this geometry, the field gradient would allow an NMR resonance to occur only in a thin horizontal layer, whose position could be moved continuously either up or down by sweeping the NMR frequency, allowing us to obtain a true 1D profile of the magnetization in the NMR region.

On April 7, I once again started putting the cryostat back together, however at about midnight, as I tightened the bolts which held the liquid helium dewar in place, the dewar suddenly exploded, sending a shower of glass into the pit below. The only spare dewar we had was too short for my long inner vacuum can, and I was lucky to notice this before I broke it as well! After machining a spacer to lower the position of the helium dewar, I again put up the dewars, but then had problems with the pressure transducer.

I began to really feel the pressure at this point. Our Physical Review Letter had just come out, and people were beginning to criticize our interpretation that the transition in the solid was first order. In addition, John Goodkind and Victor Vvedensky had both suggested that the pressure signature of the A transition was consistent with a BCS transition in the liquid. While this interpretation could not explain the high melting pressures we had measured (which suggested more entropy in the solid at low temperatures than one would expect based on a phase transition at 2 mK), I felt we needed to make a definitive NMR test of our model, and soon!

I finally succeeded in cooling the cryostat back down on April 10, and on April 14 started a compression to see if Michael Fisher's ideas regarding solid growth were correct. I was nervous as I began to form solid. With great relief, we soon found that Michael was exactly right! We almost always saw only two or three solid growth sites within the NMR region, with an all-liquid signal between the solid peaks.

I studied the solid growth characteristics throughout the middle of April. Much to my relief, the increase in growth rate of the solid peak heights as we cooled through the A transition was equally evident in all of our now spatially differentiated solid peaks. In addition, however, there was a curious, though small, drop in the solid peak heights which correlated with the backstep in pressure seen at the B transition, typically 2–3% of the total peak height. In these runs, we would decrease the gain on our chart recorder to keep the ever-growing solid peak heights on scale, but on the night of April 20 my eyes were attracted to the tiny liquid signal between the solid peaks in the data of April 17, reproduced in Figure 7.

What I saw in the liquid at the B transition hit me like a bolt of lightning: While the solid NMR signals seemed to respond to the B transition with a very small fractional change in peak height, the liquid signal almost completely disappeared at this point! Figure 8 shows the entry in my lab book that night: "2:40 am: Have discovered the BCS transition in liquid ^{3}He tonight. The pressure phenomena associated with B and B$'$ are accompanied by changes in the ^{3}He susceptibility both on and off the peaks approximately equal to the entire liquid susceptibility." I checked all the other data I had taken, and then I looked around for someone with whom to share my good news. No one was anywhere to be found in the entire building. At 4:00 am, I decided to call Dave Lee and Bob Richardson, perhaps a risky move for any graduate student. Both agreed that the identification was a strong one, and at 6:00 am Dave called back for more details.

It was now almost the time of the April American Physical Society Meeting in Washington D.C., and Dave and Bob arranged for me to give a post-deadline invited talk on our work at that meeting. We still believed that the A tran-

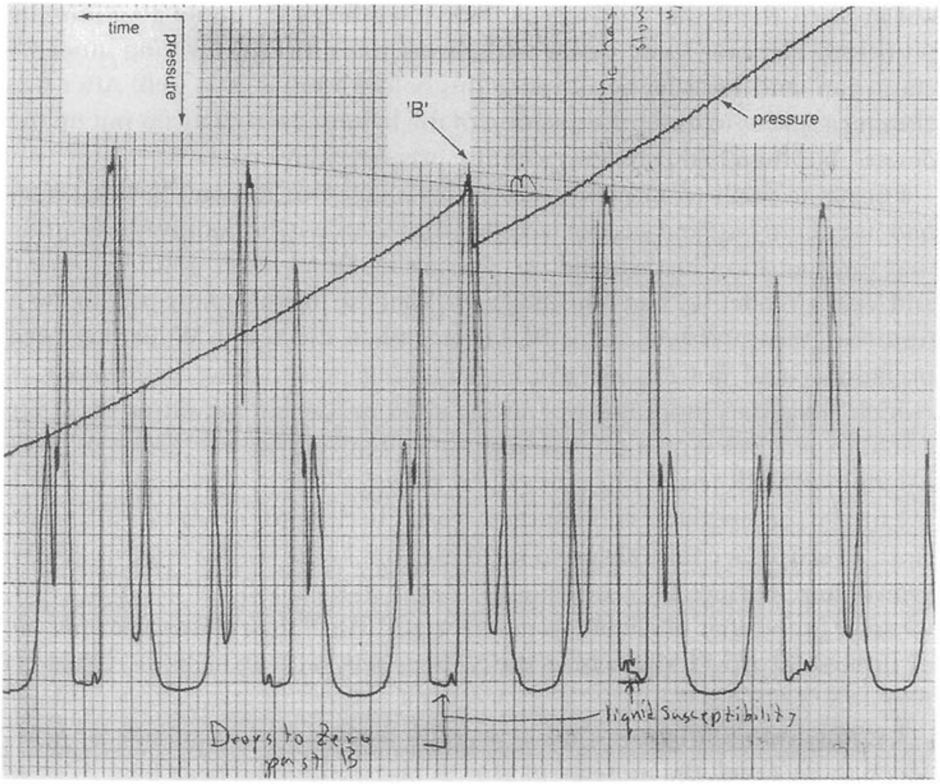
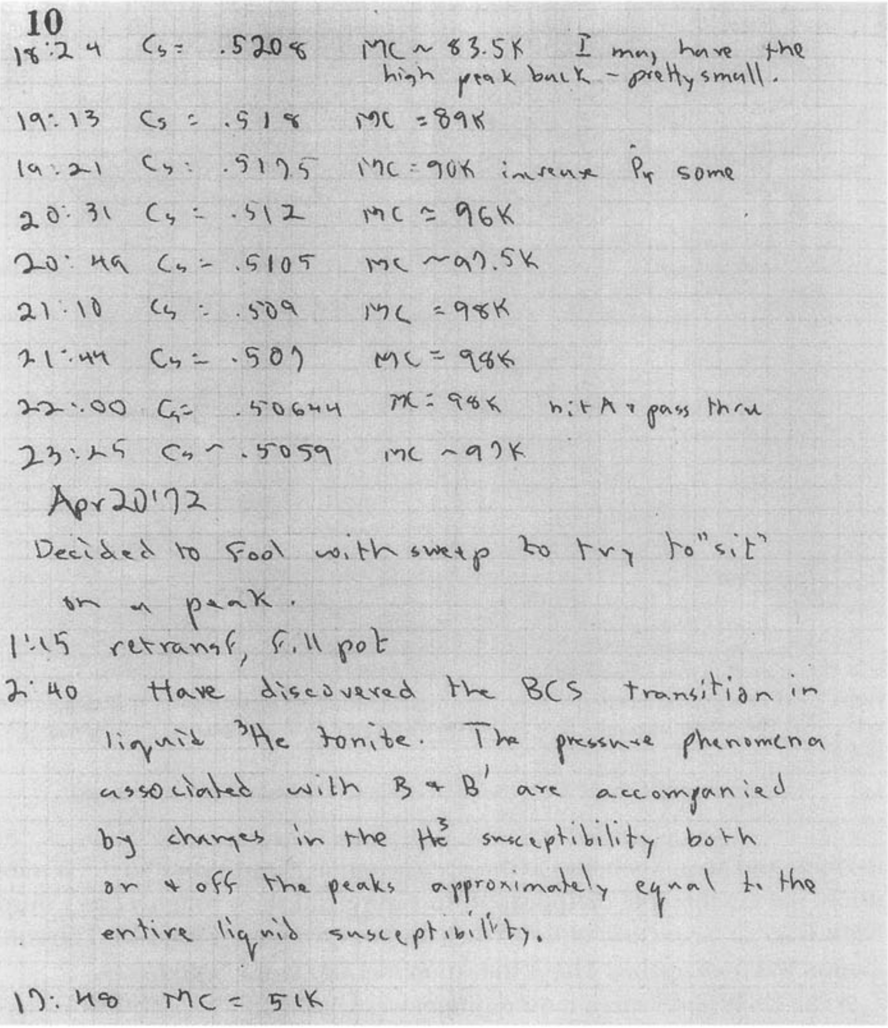


Figure 7. Continuous NMR sweeps of the cell showing behavior of the solid susceptibility (peaks) and liquid (region between two highest peaks) through the B transition. Note the subtle change in the liquid signal before and after the B transition.

sition was in the solid, so I reported that we had discovered transitions in both solid and liquid ^{3}He. When I returned from Washington, I began to study with renewed interest the supercooling properties of the B transition, and once again handed off the NMR magnet to Jim Kelly. In the meantime, I developed a multiplexer which would allow me to look at the NMR signal from two different places in the cell almost simultaneously.

When I got the magnet back in early May, I found using the multiplexer that the B transition occurred at the bottom of the cell first, and that the A-B interface moved upward at a velocity of many cm/sec. We also found a very strange magnetic signature as the A-B interface moved through the NMR resonance region: The NMR absorption signal would first rise *above* the level of the normal liquid signal, and then drop to well below that level. This effect seemed extremely difficult to understand, and we eventually tried to get the theorists down from the 5th floor in hopes of getting an explanation by offering beer and popcorn. They came, ate the popcorn, drank the beer; and then shook their heads and departed.

Beginning to feel the potential importance of these experiments, I decided to make motion pictures of the data we were obtaining in real time. Those reels of film still lie, largely unseen, in a chest in my laundry room at home. I



10
18:24 Cs = .5208 MC ~ 83.5K I may have the high peak back - pretty small.
19:13 Cs = .518 MC = 89K
19:21 Cs = .5175 MC = 90K increase $\dot{P}_T$ some
20:31 Cs = .512 MC ≃ 96K
20:49 Cs ~ .5105 MC ~ 97.5K
21:10 Cs = .509 MC = 98K
21:44 Cs = .507 MC = 98K
22:00 Cs = .50644 MC = 98K hit A & pass thru
23:45 Cs ~ .5059 MC ~ 97K
Apr 20 '72
Decided to fool with sweep to try to "sit" on a peak.
1:15 retransf, fill pot
2:40 Have discovered the BCS transition in liquid ^{3}He tonite. The pressure phenomena associated with B & B′ are accompanied by changes in the He^3 susceptibility both on & off the peaks approximately equal to the entire liquid susceptibility.
17:48 MC = 51K

Figure 8: Photograph from my lab book showing entry the night of April 20, 1972, when I realized the B transition was in the liquid.

also took a portrait of myself next to the cryostat. I reproduce that photograph in Figure 9. The haggard expression on my face was quite genuine.

The final revelation in our odyssey came sometime near the beginning of June. Curiously, nothing is mentioned in the lab book. Dave Lee encouraged me to remove the iron shims from between the NMR magnet pole faces to eliminate the gradient in the magnetic field. He wanted to test for an NMR frequency shift such as one could get in a magnetically ordered system. Indeed, we had already seen 'distortions' of the liquid ^{3}He NMR profile with the gradient applied. Both Dave and Bob were there as I cooled through the A transition. What we saw was almost too much for words: As we cooled below the A transition, a small satellite line shifted gradually to higher and higher frequencies above the larger solid peak. It resembled the all-liquid signal in both

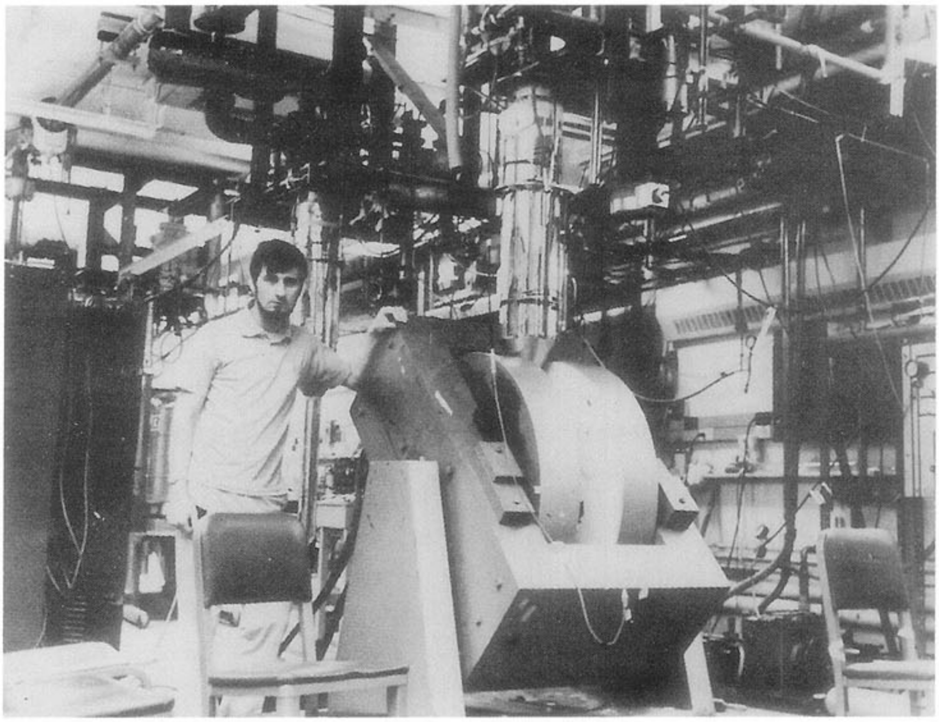


Figure 9: Self-photograph of myself taken some time in April, 1972 with my left hand on the NMR magnet used in our work. The cryostat, suspended from above, is inside the glass dewar seen entering the magnet field region.

its shape and area. Then, just as the pressurization curve indicated the B transition, the satellite line disappeared! In Figure 10 I show roughly every third NMR trace as a function of time from the compression of June 13. The conclusion was inescapable: The A transition was also in the liquid.

Willie Gully and I spent most of June investigating this unprecedented frequency shift in the liquid. At a suggestion by Bob Silsbee, we found that it obeyed what Dave Lee termed a 'Pythagorean' relationship: $(\nu_{\text{liquid}})^2 - (\nu_{\text{larmor}})^2$ was found to be a function only of temperature. Here ν_{larmor} is the precession frequency of the spins in the normal state. This difference rose from zero at the A transition to about 10^{10} Hz^2 at the lowest temperatures attainable. Vinay Ambegaokar assured us that one could not get such a shift from any conventional BCS transition. He was right. On July 14 we terminated the run and I began writing my thesis rather feverishly as Willie Gully began to modify the Pomeranchuk cell to include a vibrating wire viscometer.

Aware of how important our new understanding of the A and B features were, particularly in light of our previous published erroneous interpretations, we quickly wrote up a new manuscript and submitted it to Physical Review Letters in early July. Having learned our lesson, and certainly not able to understand the frequency shifts we had discovered, we simply focused on presenting the data, and avoided ever suggesting that these might be superfluid transitions. Ironically, although the earlier manuscript had sailed

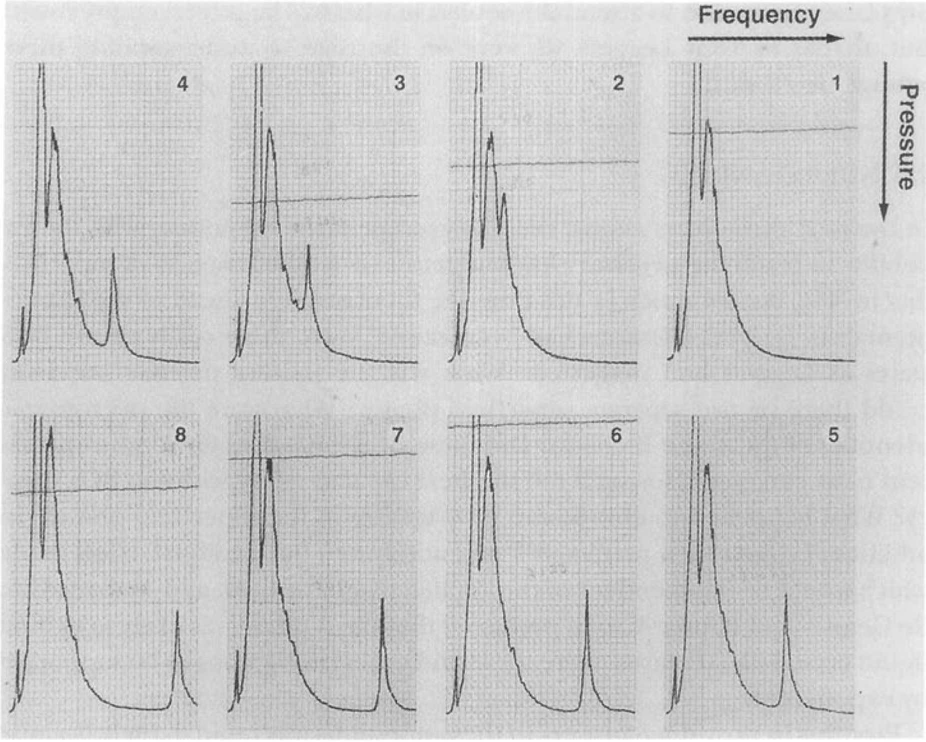


Figure 10: Sequence of NMR traces without applied magnetic field gradient as the temperature is slowly decreased below the A transition. As the liquid cools, a satellite NMR line can be seen to shift to higher frequencies. The nearly horizontal line in the traces is the cell pressure, increasing slowly from trace 1 to 8. Between traces 3 and 4 the capacitance bridge was rebalanced.

through the review process, this manuscript, in which we had much more to present, and had been very careful in doing so, was rejected. As the referee stated: "I read very carefully the previous Letter (PRL 28, 885 (1972)) and compared it with the contents of the present letter. Although the letter is clearly written, and, I presume, gives proper credit to others, I think the difference in the results is not large enough to warrant fast publication, in particular if one takes into account your rule against serial publication." Eventually, after both editors upheld the referee's conclusion, Jim Krumhansl, an associate editor of Physical Review Letters, interceded on our behalf, and the paper was published12).

In August 1972 the 13th International Conference on Low Temperature Physics was held in Boulder, Colorado. I attended the conference before reporting for work at Bell Laboratories. David Lee presented our results in a plenary invited talk. John Wheatley, who was as fast to check on our results as we had been to check on his, also spoke with supporting evidence. But the most spectacular talk of the conference, for me at least, was one by Tony Leggett, read by his colleague Mike Richards. Tony showed how our NMR frequency shifts could be produced by a p-wave BCS state in the liquid. My own talk came on the last afternoon of the conference, and even I had to change my plane reservation to attend the session in what was largely an empty room! But, thanks to Tony Leggett, we were on the road to understanding these strange new fluids.

2. UNDERSTANDING

In the next three years almost every low temperature laboratory with the capability to reach the necessary low temperatures studied aspects of superfluidity in ^{3}He, but for much of this time the theorists were ahead of the experimentalists. Several questions had been raised: Were these really p-wave BCS states as Leggett had suggested? What was the pairing mechanism? How could there be two separate superfluid phases? What were the microscopic identities of the A and B phases? Did these BCS states actually support persistent mass currents? How well did the heat capacity agree with the BCS theory? What happened to ultra-sound propagation in the superfluid phases? In addition, Leggett soon predicted13) an entirely new 'longitudinal' NMR mode which would be independent of the applied magnetic field, and Ambegaokar, de Gennes and Rainer14) soon predicted that the A phase was likely to exhibit liquid crystal-like textures. Were these and other predictions to be supported by experiment?

Progress in providing answers to these questions was rapid, largely because so much theory and experiment existed from which to draw. In the 1960s John Wheatley and others had studied the normal state properties of liquid ^{3}He extensively15). In 1961 Anderson and Morel16) had discussed a manifold of possible p-wave BCS states which had a curious property that the angular momenta of all the Cooper pairs pointed in a single direction in space, and formed only with parallel spin orientations. This state was ultimately identified with superfluid ^{3}He A. In 1963, Balian and Werthamer17) had shown that within the entire manifold of p-wave states, the state with the lowest free energy within the weak coupling BCS limit was a state in which the Cooper pairs formed with orbital angular momentum $L = 1$, spin angular momentum $S = 1$, but total angular momentum $J = 0$. A variation of this state was ultimately identified with superfluid ^{3}He B, and the formalism which Balian and Werthamer developed was adopted by everyone entering the field. In 1965 Leggett18) had studied the expected magnetic susceptibility of the nuclei in this phase, including Fermi liquid corrections. In 1971, Layzer and Fay19) had shown how ferromagnetic spin fluctuations in liquid ^{3}He could lead to an attractive interaction between quasiparticles for odd-l pairing, which they predicted would lead to superfluidity in a p-wave state. This paper provided the correct pairing mechanism for the p-wave superfluids as we understand it.

The question of how there could be two separate superfluid phases was answered very elegantly by Anderson and Brinkman20), who assumed that ferromagnetic spin fluctuations in the liquid produced the attractive interactions leading to Cooper pair formation, but noted that in ^{3}He, *but not in conventional superconductors*, the formation of the condensate wave function would modify this pairing interaction. The two concluded that the A phase should be an Anderson-Morel state which has been termed the Anderson-Brinkman-Morel, or ABM state; and that the B phase should be the Balian-Werthamer, or BW state.

At Bell Laboratories I was given an empty lab and enough money to fill it with whatever equipment I needed to continue my studies of the new phases of ^{3}He. In addition, I was allowed to hire Wolfgang Sprenger, an excellent technician who had been working for Robert Pohl at Cornell. However, the lab space I had been given was occupied by one of Bernd Matthias's people, who did not want to move out until his new lab was completed, and seemed in no rush to complete the design work. Matthias had so much influence at Bell Labs, even after leaving for U.C. San Diego, that there was nothing I could do but wait. Even so, by July 1973 my dilution refrigerator was installed and operational, thanks largely to Sprenger's efforts. By September I had reached the A transition in my new Pomeranchuk cell.

My main interest was in establishing the microscopic identities of the A and B phases. Anderson and Brinkman had suggested possible state identifications, but these identifications needed experimental verification. I also wanted to investigate the narrow region formed when the A transition split in the presence of an intense magnetic field, but needed a high-homogeneity superconducting magnet to do so. It would take one and a half years for this magnet to be delivered. There seemed only two ways for me to proceed: The first was to measure accurately the NMR susceptibility of the B phase and compare it with the 1965 Leggett prediction. The second was to search for the longitudinal NMR resonance which Leggett13) had predicted and compare it to the NMR shifts we had seen at Cornell in the A phase.

To understand the spin dynamics of p-wave superfluids, one must realize that the formation of Cooper pairs prevents the net nuclear dipole-dipole energy from averaging to zero, due to what Leggett termed a 'spontaneously broken spin-orbit symmetry'. To understand this, consider a fictitious Cooper pair in which the atoms actually orbit about one another as shown in Figure 11a. The dipole-dipole energy will be minimized over one period of the orbit when the projection of the total spin along the direction of the orbital angular momentum is zero. This spin direction is usually referred to as **d**, and in equilibrium **d** should point parallel to the orbital angular momentum of the Cooper pair. In the ABM state, the angular momenta of all the Cooper pairs are oriented in the same direction locally, a direction which we call **l**. Thus in the A phase one might expect that in equilibrium **d** would point parallel to **l**. Since the spin projection along **d** is zero, the Zeeman energy will be minimized when **d** is perpendicular to the static magnetic field, **B**. This forces both **d** and **l** to orient perpendicular to **B** in bulk A liquid21).

In an NMR experiment, one applies a magnetic field oscillating at the precession frequency of the nuclear spins in the static magnetic field, **B**, which will cause the magnetization to tip away from **B** and to precess about it. This disequilibrium perturbs the spin system, causing **d** to oscillate as shown in Figure 11b for weak oscillating fields. For conventional or 'transverse' NMR where the oscillating field lies in the plane normal to **B**, **d** oscillates in the

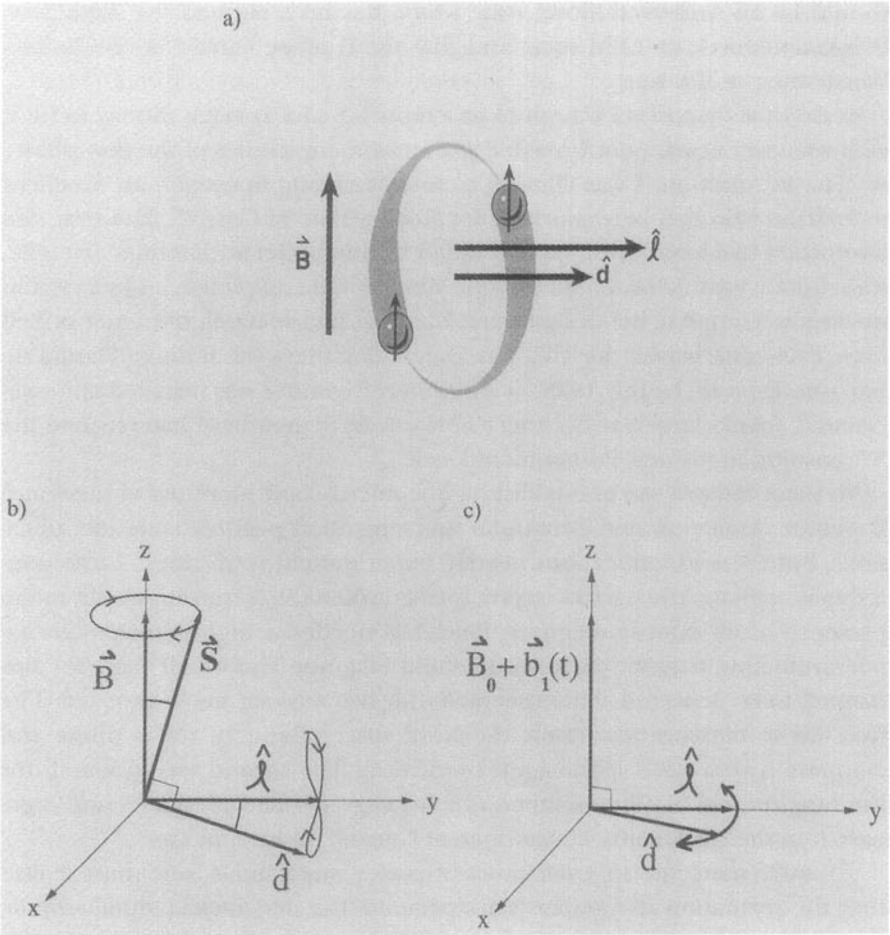


Figure 11: Superfluid A phase spin dynamics: a) A fictitious Cooper pair. The dipole energy of the two orbiting nuclei is minimized when the component of total spin, $m_s = 0$ along the orbital angular momentum, **l**. b) In transverse resonance, the magnetization, **S**, precesses about the magnetic field **B**, causing **d** to trace out a shallow figure '8' nearly in the **B-l** plane. c) When the oscillating field $\mathbf{b}_1(t)$ is parallel to the static field, the disequilibrium in the magnetization oscillates along the z-axis, causing **d** to oscillate back and forth in the x-y plane.

plane containing both **l** and **B**. However, if one applies an oscillating field along **B**, then **d** tends to rotate in the plane normal to **B** as shown in Figure 11c. This is what should happen in longitudinal resonance. When **d** is driven away from its equilibrium orientation, it exerts a torque on the precessing magnetization for transverse resonance, whose net effect is to produce the transverse resonance shift. For longitudinal resonance, the oscillating **d** causes the magnetization along **B** to oscillate in magnitude.

The longitudinal resonance was expected to be a difficult thing to find, for its frequency would be independent of any applied external magnetic field. Thus one could not search for it by sweeping the field. There was some speculation that the resonance might be extremely narrow. If the A phase were the ABM state, then $(\nu_{\text{longitudinal}})^2 = (\nu_{\text{liquid}})^2 - (\nu_{\text{larmor}})^2$, where ν_{liquid} is the conventional, or transverse, resonance we had measured in the

A phase. This suggested a value which varied from zero to about 100 kHz at the lowest temperatures. I searched hard and long for such a mode, but found nothing. In the end, I gave up the idea that the mode would be sharp, and assumed instead that it would be very broad. In this case, one could not detect it by sweeping the NMR frequency, since one would then sweep over the broad resonance of the NMR tank circuit. I decided to hold the frequency fixed and sweep the temperature, thereby sweeping the longitudinal resonance through my probe frequency. This strategy worked quite well, and the resonance was very broad. These results22) seemed to confirm that the A phase was indeed the ABM state, and soon the Cornell group23) joined in the study of this new resonance mode.

It turned out to be more difficult to identify the B phase with the Balian-Werthamer (BW) state. The B phase was a quasi-isotropic state consisting of J=0 Cooper pairs, except that as Leggett21) had showed, to minimize the nuclear dipole-dipole energy for this state, one had to rotate the spin coordinate system in which the pairs had been formed with respect to the orbital coordinate system about an arbitrary axis, **n**, by an angle of $\cos^{-1}(-1/4) \simeq 104°$. In this state, one third of the Cooper pairs should be in a $m_S = 0$ state with respect to the applied magnetic field, and ignoring interactions in the Fermi liquid, one would thus expect a susceptibility, χ_B, at $T = 0$ of 2/3 that of the normal state liquid. The Fermi liquid corrections which Leggett18) had made in 1965 reduced this value to only about 1/3 that of the normal state.

I went back and looked at the NMR absorption values I could glean from my old data, and concluded that χ_B was at least close to that which one would expect for the BW state. However, very soon Wheatley's group at U.C. San Diego (La Jolla) measured the B phase static magnetization using an RF SQUID magnetometer, and found that the drop in magnetization was considerably larger than expected for the BW state24). Later, Lounasmaa's group in Helsinki reported25) an NMR measurement of χ_B which also showed a larger drop that expected. There were suggestions that perhaps the B phase might be a mixed p and d-wave state.

When I began to look at the B phase NMR susceptibility, I found a very strange thing: If I measured it in a low magnetic field, I would find a very low value relative to the normal phase, while if I measured it in a high magnetic field, I would find a value much closer to the Leggett prediction for the BW state. In addition, I found that there was always a high-frequency tail on the B phase NMR line. This is precisely the effect which plagued the early Helsinki measurements.

At this point I enlisted the help of Bill Brinkman in order to understand my strange observations. Within the Leggett theory, the NMR frequency in the BW state should depend upon how the rotation axis, **n**, is oriented with respect to **B**. There should be a shift proportional to $\sin^2(\theta)$, where θ is the angle between **B** and **n**. In the bulk, **n** should orient parallel to **B** to re-minimize the dipole energy as a result of depairing of the spin component for which $m_S = 0$ along the magnetic field21). Thus, in the bulk, the BW phase should show no NMR frequency shift. However, if surfaces favored a different orientation, there could be a variation of orientation across the sample, which would result in a broad high frequency tail on the NMR line, with a resulting decrease in the spectral weight at the Larmor frequency. This might help explain my observations of NMR in the B phase. We were able to determine a lower bound for the B phase NMR susceptibility by going to high fields (about 0.1 T), however, and this limit was much closer to the predicted values for the BW state than to the La Jolla value based on static magnetization measurements[22)].

I and others were to make successively better and better measurements of the B phase NMR susceptibility[26, 27)], and Wheatley and others[28)] were to make better and better measurements of the static magnetization. They never came into agreement. While it is fairly universally agreed that the B phase is the BW state, and that the NMR susceptibility measurements support this, there seems to be a change in static magnetization at the A-B transition which exceeds the nuclear spin contribution, and which has never been understood.

By 1974 Pierre de Gennes[29)] had already described how one should carry the idea of Ginzburg-Landau bending energies over from liquid crystals to the ABM state to describe liquid-crystal-like textures in the A phase. Brinkman, Smith, and Blount[30)] produced a similar description for the BW state to try to understand my early NMR experiments in the B phase. They were able to show that within the cylindrical geometry used in my experiments, their theory fit the data, provided only that one assume that the equilibrium orientation of **n** near the cylindrical walls was not parallel to the magnetic field (which was oriented along the cylinder axis). This was a triumph for us, because it would be quite some time before these ideas could be tested in the A phase, owing to much stronger orienting effects in that phase.

Later experiments at Bell Labs and in Helsinki were to demonstrate the specific orienting effects by surfaces on the B phase textures[31,32)], and at Bell Labs we would observe spin-wave modes trapped in a B phase texture which existed between parallel surfaces[33)], while the Helsinki group would detect a spin mode associated with spin-waves trapped on quantized vortices in the A phase[34)]. The frequency shifts of the B liquid between closely spaced plates resulting from the surface orientation of **n** suggested B phase longitudinal resonance frequencies which agreed well with direct measurements of the B phase longitudinal resonant frequency[35)], which in turn agreed well with values calculated from the A phase longitudinal resonant frequencies using relationships due to Leggett.

During most of this period, the Helsinki group was the only group to be using copper adiabatic nuclear demagnetization for refrigeration. They were thus the only group which could observe superfluidity in ^{3}He at zero pressure, and were quick to do so, extending the phase diagram determined by the La Jolla group to zero pressure. The superfluid transition temperature diminished from about 2.5 mK at melting pressure to 0.9 mK at zero pressure, while the equilibrium B′ transition temperature rose nearly linearly with decreasing pressure, until below about 21 Bars only the B phase was stable in low magnetic fields. The La Jolla and Helsinki groups both made measurements of the heat capacity of the liquid through the superfluid transition, and found that the superfluid looked very much like a weak coupling BCS state at low pressures, but strong coupling effects at high pressures caused the gap in the A phase to open up faster than at lower pressures. In addition, the Helsinki group extended many of my NMR measurements along the melting curve to lower pressures36).

In late 1974 my high-homogeneity NMR magnet finally arrived, and I decided to look closely at the tiny region called the A_1 phase that existed between the split A transition in a magnetic field. It was presumed that in this region only one of the two spin species had undergone pairing, but it was not known which. When I had planned the development of my lab, I had been concerned that by the time I got running, the *only* phase left to study would be the A_1 phase. I wanted to measure the slope of the frequency shift vs. temperature in this phase, $d(\nu_{\text{liquid}}^2 - \nu_{\text{larmor}}^2)/dT$, and compare it to the equivalent slope in the A phase at low fields. This ended up being a formidable task, however, because the maximum shift in the A_1 region turned out to be only about 3 Hz, independent of magnetic field. As usual, I was lucky, and found that owing to copper foils I had used to shield the oscillating NMR field from solid ^{3}He in my cell, a portion of my NMR line was extremely narrow, allowing frequency shifts to be measured to 0.1 Hz, even at an NMR frequency of 24 MHz. In the end, I found the ratio of the slopes to be 0.188, but I had no idea what this number meant.

I enlisted the help of Phil Anderson to interpret the A_1 phase data. He worked out the theory in a Sunday afternoon. His theory not only showed that the ratio stated above was consistent with the Anderson-Brinkman identifications of the A and B phases as the ABM and BW states, but correctly predicted the shape of the curves I had measured37). Unfortunately, this combination of theory and measurement still did not indicate which spin species had undergone pairing in the A_1 phase, and while there is some evidence today that it is the spins parallel to the magnetic field, the evidence is not entirely convincing38, 39). Ironically, the most important contribution from this early work on the A_1 phase was probably not in the detailed measurements, but in the simple fact that we actually saw a transverse shift at all. In 1975 David Mermin40) was to show that the most likely $L = 3$ (f-wave pairing) candidate for the A_1 phase would not have a transverse shift. This is probably still the best evidence that the A and B phases could not be f-wave states.

By mid-1974, there were at least four groups studying the new phases. Groups at Cornell, La Jolla and Argonne had found that they could excite normal modes of the superfluid order parameters with ultrasonic sound waves, and this technique provided another incisive probe of the superfluids. The Cornell group later found very interesting non-linear response to sound waves, while the La Jolla group had studied linear and non-linear ‘ringing’ of the magnetization in the superfluid phases when a small magnetic field was abruptly turned off.

In the summer of 1974 Linton Corruccini, with whom I had worked at Cornell as a graduate student, came to Bell Labs and we had a wonderfully productive summer. Henrik Smith was also there from Copenhagen, and he and Brinkman were working together on the theory of superfluid ^{3}He. Corruccini and I decided to try some pulsed NMR experiments, which had never been used to study the superfluid phases. It was necessary to completely shield the solid ^{3}He which formed as we compressed from the oscillating magnetic field, and we created a very clever design to accomplish this.

Corruccini and I first measured the B phase magnetic susceptibility with pulsed NMR27), and got extremely good agreement with my previous continuous wave NMR results. We then set out to study spin dynamics in which the magnetization was tipped far from equilibrium. Brinkman and Smith predicted how the A phase frequency shift should depend upon tipping angle, and we found their prediction to describe the behavior we observed remarkably well41). However, when we asked them to predict what we would see in the B phase, they weren't even close. As we increased the angle by which we tipped the magnetization away from the static field, no frequency shift was observed at all, until we reached an angle of about $(104 \pm 1.5)^\circ$, at which point a rapidly increasing frequency shift was observed42). This angle was remarkably close to the angle by which one had to rotate the spin coordinate system with respect to the orbital coordinate system to minimize the dipole energy in the BW state, and Bob Richardson was elated to see a direct measurement of this angle. Brinkman and Smith43) were able to determine what the spin system was doing in our experiments, and it became clear that we had indeed made a direct measurement of the rotation angle. But the two could never really understand why the spin system behaved as it did. This understanding was finally supplied by a Russian theorist Igor Fomin44). Corruccini and I also made the first studies of longitudinal spin relaxation in both superfluid phases45). In all, work that summer resulted in seven publications, including three Physical Review Letters.

In the course of our pulsed NMR experiments, Corruccini and I observed in the B phase that, for sufficiently large tipping angles, the tipped magnetization would precess coherently for times much longer than should be possible, based on the magnetic field gradients which existed across our sample. We called this behavior a 'zero k spin-wave' but never really understood it, and its existence was buried in conference proceedings42,46). Years later, Bunkov and co-workers in Russia were to re-discover this phenomenon47), and Fomin explained the behavior48) as being due to spin supercurrents in the B liquid driven by gradients in the order parameter as the magnetization in the regions of field gradient wound up in a helix. These spin supercurrents effectively increased the tipping angle in regions of lower magnetic field. Once the tipping angle exceeded 104°, the NMR frequency in that portion of the sample would begin to increase. Once the frequency there became equal to that in regions of higher magnetic field, the spin supercurrents would cease, and the entire sample would precess in-phase. They named this beautifully orchestrated dance the 'homogeneously precessing domain', and used it to study many unusual phenomena in the B superfluid.

There is not enough space nor time to complete even this abbreviated account of our growing understanding of these remarkable fluids. I encourage the reader to learn more in a series of excellent review articles written during this period of discovery by some of its most important contributors.49) I will mention briefly in closing, however, what happened to the search for anti-ferromagnetism in solid ^{3}He; the topic which Dave Lee had assigned for my Ph.D. thesis.

In 1974 Bill Halperin50), one of Bob Richardson's graduate students, did use Pomeranchuk cooling to observe the drop in spin entropy which signaled nuclear ordering in solid ^{3}He, but not at 2 mK as had been expected based on measurements above T_C, but at about 1 mK. In addition, the transition was strongly first order, rather than second order as had been expected. Halperin developed a clever self-consistent technique for extending the melting curve measurements to below the solid ordering temperature, which did not rely on any secondary thermometer. Later, Dwight Adams' group at the University of Florida51) was to apply this technique in high magnetic fields, and in the process discovered that above about 0.4 T there was a second magnetically ordered solid phase with a much higher magnetization than the low field phase. The nature of the magnetic order in this high field phase was first guessed by theorists52), however the nature of the low field phase remained a mystery.

Finally, in 1979, I again began to think about the solid, as did Dwight Adams at the University of Florida. Borrowing and modifying a strategy which I had first heard mentioned by Bill Halperin, and using copper nuclear demagnetization for refrigeration, I learned to grow single crystals of solid ^{3}He directly from the superfluid into the magnetically ordered solid phase. Magnetic resonance experiments at both Florida53) and Bell Labs54) then showed a rich NMR spectrum in the low field ordered phase, immediately showing that the sublattice structure had a symmetry lower than cubic. Mike Cross, Daniel Fisher and I were able to determine the symmetry of the ordered state from our NMR spectra, and to ultimately guess the exact sublattice structure. We found that the low field phase consisted of ferromagnetic planes of spins normal to any one of the principal lattice directions, with the spin orientations in these planes alternating two planes up, then two planes down, etc.55) This work was done at about the time that the movie 'Star Wars' was being shown, and in honor of the robot R2D2 in that movie, we named the ordered phase 'U2D2' meaning 'up 2, down 2'.

In solid ^{3}He the atom-atom exchange energy, which produces the spin order, is nearly four orders of magnitude larger than the direct nuclear magnetic dipole-dipole energy, and four orders of magnitude smaller than the characteristic lattice energy, the Debye energy. This has made these magnetic systems particularly convenient model systems for studying cooperative magnetic behavior56). Without question we have been very fortunate that, within three thousandths of a degree of absolute zero, there exist a total of five beautifully ordered phases of solid and liquid ^{3}He, whose behavior continues to challenge our understanding and to provide a proving ground for new ideas.

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