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OBSERVATION OF STRONGLY POLARIZED LIQUID AND SOLID ^3He USING THE CASTAING-NOZIERES EFFECT AND POMERANCHUK COOLING

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Résumé - L'Helium trois étant un liquide de Fermi, pour obtenir des polarisations proches de 1 par brute-force il faut appliquer un champ magnétique $H \sim K_B T_F / \mu \sim 200$ T trop grand pour être maintenu en laboratoire pendant un temps suffisamment long. Castaing et Nozières ont suggéré une méthode très élégante qui ne fait pas appel à des champs aussi élevés. Il s'agit de polariser ^3He solide, qui est en première approximation un paramagnétique de type Curie-Weiss, et de le liquéfier en un temps court devant T_1 , le temps de relaxation de l'aimantation. Nous avons observé le bien fondé de leur hypothèse, l'aimantation étant maintenue dans le liquide avec un temps de relaxation de quelques minutes. Des polarisations de solide d'environ 78% ont été obtenues récemment. A cause des interactions antiferromagnétiques il faudra des champs magnétiques de l'ordre de 14 T pour obtenir des polarisations proches de 100%. La plupart de l'aimantation du solide peut être transférée en liquide.

Abstract - Because of the Fermi character of liquid ^3He , to polarize this system to values close to one by brute-force we should apply a field $H \sim K_B T_F / \mu \sim 200$ T far too high to be produced in the laboratory for a sufficiently long time. Castaing and Nozières suggested a very elegant method of overcoming this problem which consists in polarizing solid ^3He (which has a magnetic susceptibility of the Curie-Weiss type) and quickly melt it in a time short compared to T_1 , the time for the magnetization to relax. The magnetization was observed to stay in the liquid phase with a relaxation time of a few minutes. Using Pomeranchuk cooling, solid polarizations as high as 78% were recently obtained. Because of antiferromagnetic interactions, larger solid polarizations will need higher magnetic fields. Most of the solid magnetizations can be transferred to the liquid.

1. Introduction - At the present state of technology it is impossible to achieve liquid ^3He magnetizations m_L much larger than 10% of the saturation magnetization m_S , by brute-force, that is cooling under a high magnetic field.

The condition to achieve $m_L = m_S$ implies $\mu H = K_B T_F^*$ where $\frac{\mu}{K_B} = 0.8$ mK/T and $T_F^* = 179$ mK at $P = 33$ bar, (1) which means $H = 225$ T a field about ten times larger than what can be sustained at present with the best Bitter magnets.

One possible way would be polarizing ^3He gas by optical pumping and then condensing it. Very good results have recently been obtained in polarizing the gas by M. Leduc and F. Lalöe (2) using a laser of the convenient frequency. Although interesting properties are expected from the gas phase (3,4) liquefaction and conservation of the polariz-

ation will probably still demand a lot of experimental effort.

A totally different approach was suggested by B. Castaing and P. Nozières (5). Their idea was to use strongly polarized solid ^3He which to a first approximation they supposed a simple Curie paramagnet (polarization would be about 90% at 4 mK and 7 T). If solid could be melted in a time short compared to T_1 , the relaxation time of the magnetization, then the liquid should conserve the magnetization of the solid.

We would then have

$$m_S = \chi H_{\text{ext}} \approx \frac{C H_{\text{ext}}}{T}$$

$$m_L = \chi H \approx \frac{C H}{T_F^*}$$

if $m_S = m_L$ then $H = H_{\text{ext}} \frac{T_F^*}{T}$ which, for $T = 4$ mK and $H_{\text{ext}} = 7$ T would give $H \approx 300$ T a substantial increa-

se of the applied field indeed.

Assuming we can produce and measure strongly polarized solid ^3He , the only difficulty to observe the Castaing-Nozières effect would be related to the relaxation time T_1 , this is a combination of intrinsic dipole relaxation, due to collisions between ^3He atoms, and extrinsic relaxation, at the walls of the container.

The intrinsic relaxation time is directly proportional to the spin diffusion coefficient (5,6). Extrinsic relaxation time will depend on how fast the ^3He quasiparticles can diffuse to the wall, where they will relax hence $T_1 \sim D^{-1}$. In the degenerate region liquid ^3He has a diffusion coefficient that goes as T^{-2} with a broad minimum of about $3 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$ at a few hundred millikelvin. We would then expected that at those temperatures and for relatively large volumes (a few cubic millimeters) intrinsic relaxation times of 5 or 6 minutes should be observed.

Decreasing the dimensions of the ^3He cell would lead to a predominantly extrinsic relaxation. Since coating the walls with ^4He can reduce the wall relaxation by more than a factor 10 (6,7) typical minimum distances between walls of 0.1mm could still be used without much problem.

2. The Method - To observe the Castaing-Nozières effect we used a specially designed Pomeranchuk cell for reason which will become clear in a moment. The experiment would consist in cooling liquid ^3He inside the cell by means of a dilution refrigerator in a high magnetic field. Magnetization would be followed by continuous wave nuclear resonance using a Q-meter. During solidification the signal would increase due to the formation of magnetization solid. ^3He pressure would

then be released to a value lower than the minimum of the melting curve. The NMR signal should decrease with a time constant T_1 which we expect to be roughly the intrinsic relaxation time at the temperature at which the liquid would find itself after decompression.

In a first experiment which was done at the H.C. Ørsted Institute of Copenhagen (8) at 2.7 T, liquid polarizations up to 20% were obtained. Simultaneous measurements by Schumacher et al. (9) at Grenoble using indirect cooling of solid ^3He have also shown the feasibility of the Castaing-Nozières effect. Although polarizations were only about 9% in 3.5T field, later experiments with indirect cooling led to polarizations close to 20% in a 7.3 T field. (10)

We then made new experiments at Grenoble using Pomeranchuk compressions in a 7.2 T field, but we concentrated on the properties of strongly polarized solid ^3He (11,12) since it became clear that the second (larger) cell was not ideal to study polarized liquid ^3He . Solid polarizations near 75% were then obtained.

We will discuss briefly some problems associated with solid magnetization measurements as well as the design of the cells used on each experiment and the results obtained.

3. Magnetizations measurements and Pomeranchuk cells

- F. Rasmussen (13) discusses different possibilities for cooling solid ^3He and in particular Pomeranchuk cooling. This method relies on the fact that liquid entropy, which decreases as T well below T_F is lower than the solid entropy (equal to $R \ln 2$ in the non interacting solid picture) below about 300 mK, where the melting curve has a minimum. Isentropic solidification of liquid ^3He absorbs heat and produces cold solid.

Let us first convince ourselves that Pomeranchuk cooling is better than indirect cooling when it comes to magnetize solid ^3He in a non-restricted geometry.

Fig.1 shows the entropy of solid ^3He in zero field, and in a 7.2 T field as we deduced by taking the slope of the melting curve Fig.2 measured during the Grenoble Pomeranchuk experiment.

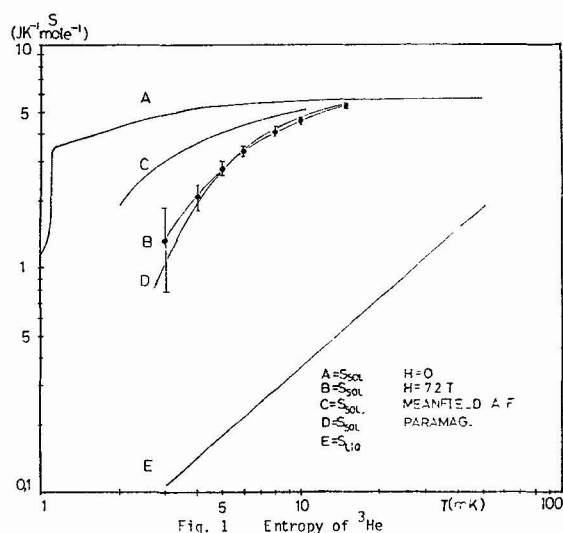


Fig. 1 Entropy of ^3He

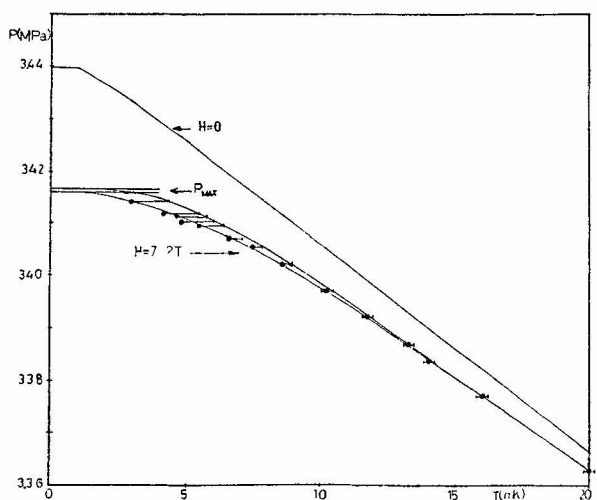


Fig.2 ^3He melting curve at $H=0$ from Ref. (28) and our results (7.2T)

We see that to cool (for example) from 10 mK to 8 mK entropy has to be reduced by about $0.6 \text{ J mole}^{-1} \text{ K}^{-1}$.

The cell described in refs. (9,10) in which 1 cm^3 of solid ^3He was indirectly cooled through a 3 cm^2 heat exchanger made of 100 insulated copper wires (coated with several layers of ^4He) would take about 100 hours to go from 10 mK to 8 mK, in the same field. (We assume a 3 mK dilution refrigerator and a reasonable Kapitza resistivity $R_K T^3 = 10^{-2} \text{ m}^2 \text{ K}^4 / \text{W}$). In practice the solid temperature was limited to $\sim 18 \text{ mK}$ implying that some other thermal resistance (or a large heat leak) appeared (14). The time constant $\tau = RC$ goes as T^{-5} due to the nuclear heat capacity term T^{-2} and the Kapitza resistance T^{-3} ; it is then very difficult to cool to much lower temperatures even if the exchange area is increased by a factor 10 or more, in which case, even if relaxation time is still long, NMR might be difficult to do.

The advantage of the Pomeranchuk cell is that we are cooling liquid ^3He which has about 13 times less specific heat (at 10 mK) and an excellent thermal conductivity allowing the use of a heat exchanger of much larger area separated from the experimental space (Fig.3)

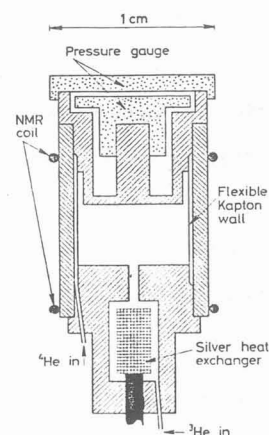


Fig.3 Pomeranchuk cell used in the Copenhagen experiment.

Solid will then be formed at temperatures at least as low as the precooled liquid. Some other difficulties might appear though, as we discuss later on.

The Pomeranchuk cells we have used have some particular features which we developed to solve some of the problems associated with precise measurements of solid ^3He magnetization particularly in high magnetic fields. Let us briefly discuss some of these problems.

The NMR aspects of the experiment and in particular the interconnection of the lattice, Zeeman and exchange reservoirs are discussed by M. Chapellier [14].

It is important to know exactly the solid fraction and the temperature distribution inside the solid formed. Using the traditional type of metallic cell [15] only part of the solid could be studied by NMR (since RF cannot penetrate the metallic container). This was usually done by adding to the cell some plastic tail around which the NMR coil was wound. Solid formation could then be induced inside the coil by means of a heater resistance or a gradient of magnetic field or pressure. The solid fraction inside the coil was the estimated but errors due to spurious and uncontrollable solid growth are far too large to derive for instance the $M \times T$ Diagram [16,17]

The situation is improved when solid is formed from the superfluid phase [18] because of suppression of thermal gradients; in fact if liquid ^3He becomes superfluid during a compression it is probable that solid will melt everywhere along the cell and resolidify at the bottom. Magnetic field distribution is also very important.

Because of the depression of the melting curve with magnetic field (Fig.2) due to solid spin en-

tropy reduction, solid will form, at a given temperature, where the field is higher. In high magnetic fields, roughly 1 to 2% liquid has to be solidified to decrease the temperature by 1 mK. Because the difficulty of cooling bulk solid it is essential that liquid be precooled to as low a temperature as possible, to avoid unwanted thermal and magnetic gradients inside the solid under study.

To complete the list of problems, thermometry under high magnetic fields is one of the most difficult, and special attention must be paid to it if quantitative measurements are to be made.

The Pomeranchuk cells that we used are then made entirely of plastic, and can be made in very small dimensions so as to fit inside a very low inductance NMR coil. All the spins can be probed in this way, and the complete cell can be placed in a highly homogeneous region of the magnet.

A precise measurement of the volume change can be made and hence the solid fraction accurately deduced as explained by Rasmussen [13].

By placing the cell inside the mixing chamber of a dilution refrigerator which can maintain 2 mK [19] continuously the hot solid fraction can be eliminated since solid can be formed directly at its nuclear ordering temperature, which we measured to be about 3.5 mK at 7.2 T [11,12,20]. Solid in high fields could in principle even be formed directly from the superfluid phase using our dilution refrigerator although we never did it (in high field) because of the small exchanger areas that we used ($\sigma \sim 500 \text{ cm}^2$). In a field of 25 mT a similar cell had been precooled to 2.8 mK essentially the temperature reached by the refrigerator at that time [21].

Their construction is as follows: A cylindrical membrane is made by wrapping a convenient number

of plastic Kapton foils (22) 12.5μ thick around a Teflon rod and gluing them together with 1266 Sty-cast (23). The ends are reinforced with double thickness of plastic foil. The membrane is cut to the desired length and removed from the Teflon rod by cooling with liquid N_2 . The epoxy end pieces (flanges) are then glued to it and a pressure test is made at N_2 temperature. The membrane is then glued inside the 4He container and holes are machined on one (or both) ends. The measuring sensors and the heat exchanger are introduced and sealed with new epoxy plugs. (Fig.4)

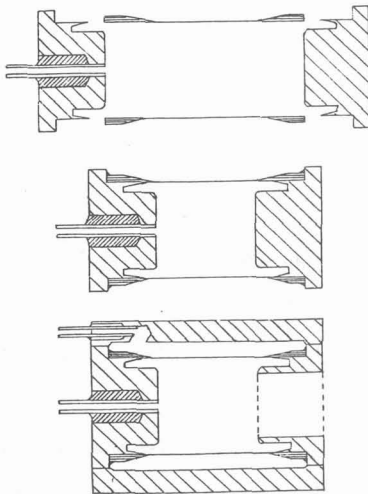


Fig. 4. Assembling a Pomeranchuk cell. For details, see text.

To calculate such cells, the procedure is as follows. We first determine the thickness of the membrane for a given radius of the cells. For cells of a few mm, since we are using Kapton foils of 12.5μ (15μ including the glue) we actually calculate the number N of layers and determine the exact radius R of the cell by means of the empirical expression

$$R = 4.4 \times 10^3 N (P_{3i} - P_{4i})^{-1} \quad (1)$$

where R is in meters and $P_{3i} - P_{4i}$ the maximum differential pressure across the membrane in Pascal.

This gives for the maximum stress

$$\sigma = 2.9 \times 10^8 \frac{N}{m^2}$$

at 77 K, which increases by about 5% at 4 K.

A long thin cylinder fixed at one end and subjected to an internal pressure P will have its radius R increased by $\Delta R(x)$ (x =distance from the end)

$$\Delta R(x) = \frac{PR^2}{Et} \{ 1 - e^{Kx} (\sin Kx + \cos Kx) \}$$

where E is the Young's modulus, t is the cylinder thickness and K is a coefficient, related to the Poisson's ratio, and equal to $k = \frac{1.28}{Et}$ for our case. The end (or flange) effect can be considered negligible when the maximum deformation $\Delta R(x)$ has reached say 90% of its maximum value $\frac{PR^2}{Et}$ which is achieved for $x \gtrsim 2 \sqrt{tR}$. For $R = 4 \text{ mm}$ and $t = 0.06 \text{ mm}$, $x \approx 1 \text{ mm}$

Since the stress at the flanges is twice that at the center, we must provide a larger thickness over the region where the effect of the flanges extends possibly decreasing with increasing x .

If we do not exceed the elastic limit, the increase in volume will be proportional to the change of the differential pressure across the membrane (usually 3He pressure inside and 4He pressure outside).

$$\Delta V = \alpha \{ (P_{3i} - P_{4i}) - (P_{3f} - P_{4f}) \} = \alpha \Delta P_{3,4}$$

from which we deduce

$$\frac{\Delta V}{V} = \frac{2R}{Et} \Delta P_{3,4} \quad (2)$$

The final pressure P_{3f} is about 3,4 MPa. Knowing that we can go from zero to 2.5 MPa on the 4He side and that the maximum differential pressure is $P_{3i} - P_{4i}$, chosen according to experimental reasons, we calculate $\frac{\Delta V}{V}$ for the cylinder defined by the membrane in the region situated at distances $x \approx 2 \sqrt{tR}$ from the end flanges.

If we take for example $P_{3i} - P_{4i} \text{ max} = 2.6 \text{ MPa}$

($P_{3i} = 3.0$ MPa and $P_{4i} = 0.4$ MPa) and $N = 4$ layers (60 meters) we deduce from (1) $R = 6.8$ mm. Using $E = 6.1 \times 10^9$ N m⁻², the Young's modulus of Kapton film at 4 K we deduce $\frac{\Delta V}{V} = 6.3\%$. This is enough to totally solidify the liquid sample, (volume decrease of about 5%) and to provide some increase of the liquid pressure (13). Roughly about 4 bars of ⁴He are needed to increase 1 bar in liquid pressure when the volume of the cell is the one defined by the membrane (ends excluded).

If the compression is to be started at a low initial ³He pressure, this fact is to be taken into account and some liquid volume has to be excluded (Fig.3). If the initial liquid temperature is of a few mK the initial ³He pressure can be set almost equal to the final pressure.

4. Experimental Results

4.1 The Copenhagen experiment: observation of the Castaing-Nozières effect - This experiment has already been described in the literature (8,24,25). We will comment on some aspects of it.

The ³He sample, which contained less than 1000 ppm ⁴He impurities was solidified in a Pomeranchuk cell whose membrane was made of 2 layers of 15 μ Kapton of 6.8 mm diameter and 8 mm total length, with the inner distance between flanges reduced to 4 mm (fig.3). The maximum differential pressure was kept below 2.8 MPa. Making use of the liquid compressibility, the solid fraction was determined through the expression

$$x_s = 5.9\% \{ \Delta P_4 - 2.5 \Delta P_3 \}.$$

The heat exchanger inside the cell was made by sintering about 50 mg of 400 Å Ag powder (area of about 900 cm²) (26) to very pure Ag plated copper foil 0.1 mm thick. About 10 times this amount extended outside the cell to provide contact with the diluted phase of the dilution refrigerator mixing

chamber.

Since we wanted a free Helium space nothing was placed inside the cell. Thermometry relied on the melting curve pressure at 2.7 T (27).

The pressure gauge of this cell was not meant for precise measurements but only as an indication. It was made by using the deformation of one of the ends of the cell, the configuration being shown in fig.3. We knew that it would be dependent on the ⁴He pressure, as was already observed in a similar pressure gauge made of much stronger BeCu (28).

A carbon thermometer was also placed inside the exchanger well, but the contact broke and we could not measure the temperature of the cell after decompression to the liquid phase. A reasonable estimate was made by recompressing back to the melting curve.

Two saddle coils were provided for NMR detection, at right angles to each other. One with 2 windings for high frequencies around 90 MHz and one with 80 windings, for frequencies of about 1 MHz. The signal was detected with a conventional Q-meter, using field sweep.

The cell was precooled to temperatures in the range 20-40 mK, usually with some solid inside. The ³He line was blocked by a solid plug, the outside pressure being kept at 3.4 MPa. The ⁴He pressure was then increased at a constant rate until a maximum signal was observed (solid fraction of 30 to 50%), when the compression was stopped and both ³He and ⁴He pressures released, in about one second.

Fig.5 shows a typical (hence one of the best) compression-decompression experiments. Each spike corresponds to an absorption curve obtained by sweeping the field.

The upper part of the figure shows schematically the pressure trace and the moment of decompression. At point D the sensitivity of the pressure gauge was

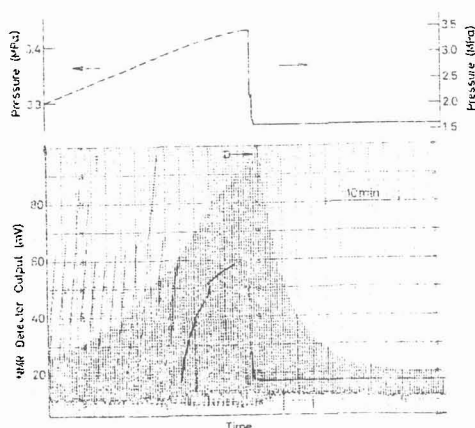


Fig.5 NMR absorption and pressure recorded during a compression of ^3He followed by a decompression.

reduced and pressure released. ^3He final pressure for this decompression was 1.6 MPa.

The NMR signal is seen to decrease almost exponentially after decompression in what we think is the liquid phase. The relaxation times as a function of final pressure and magnetic field are shown in Fig. 6.

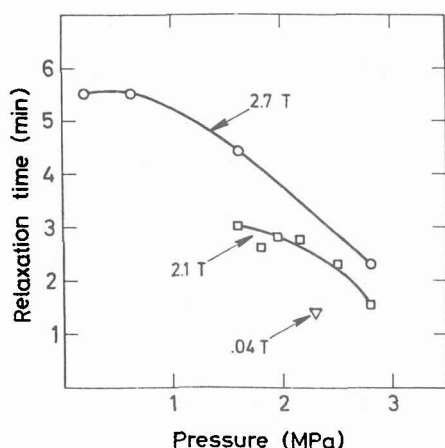


Fig. 6 - Relaxation time of polarized liquid ^3He versus the final pressure in decompressions like fig.5. The applied magnetic fields are indicated.

We could think that what we have is in fact a very expanded solid and this hypothesis cannot be totally excluded but the indications are very strong that it is actually the liquid phase. First of all there is no discontinuity in the relaxation times

which are in very good agreement with those of the normal liquid. It is hard to believe that melting of the solid will proceed at slower rates when the pressure is lower, and that it would stay solid independently of pressure.

Besides the fact that the cell pressure gauge could be actuated from the outside, indicating that there was no solid blocking the way to the gauge, we have also made some recompressions to find the melting curve. ^3He pressure was quickly increased from outside until a solid plug formed inside the ^3He capillary and ^4He pressure was then increased as in a normal compression. This method was actually tried to see the depression of the melting curve, predicted by the theory.

No significant depression was observed, but the effect expected would be very small for the polarizations achieved.

For a constant ^4He pressurization rate ^3He pressure increases with time, the $P \times t$ slope being related to the liquid compressibility.

When the melting curve is reached at a pressure P_{30} the slope decreases due to solid formation, because of the smaller molar volume of solid ^3He as compared to liquid.

If the temperature is known at P_{30} , then the melting curve can be derived and vice-versa. The existence of this change in slope is an evidence that we went from liquid to solid.

We can always argue that the solid could be overexpanded and exist even at $P = 0$ and eventually melt very slowly and have the same compressibility as that of the liquid. In this case this new "solid" would have properties just as interesting as the polarized liquid phase but then we should specify again what we mean by solid or liquid.

The liquid polarization was not accurately mea-

sured. It was deduced by taking the ratio of the signal height just after decompression, to the liquid signal after relaxation, corrected from the mixing chamber contribution (about 30% of the equilibrium liquid signal at 20 mK), and for the loss of polarization due to the increase in temperature after relaxation. Maximum average liquid polarizations of 15 to 20% were thus obtained. If we consider that only a fraction of liquid was solidified then the largest partial liquid polarizations could exceed 30 to 40%.

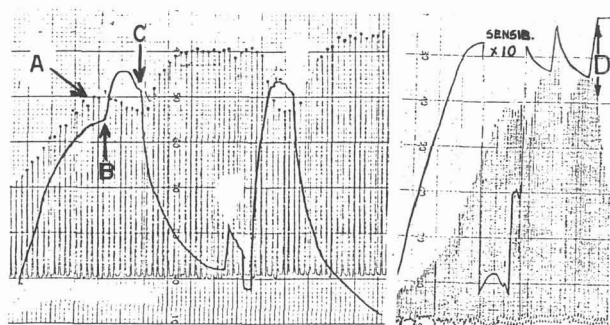


Fig. 7

Fig. 7 shows peculiar features of the NMR signals observed during some compressions.

If we kept a constant ^4He pressurization the signal increased due to the increase in solid magnetization per unit volume, (since the temperature decreased and due to the increasing solid fraction. When the pressure approached its maximum value, which is related to solid ordering and, in any case, to a decrease in solid entropy (if not a large heat leak due to solid crushing) the signal would stop growing (Point A).

Increasing the compression rate (Point B) actually increased the ^3He pressure hence producing a considerable amount of cold solid but the NMR signal actually decreased.

If the compression was halted the pressure relaxed and the NMR signal increased to values higher than at point A.

Magnetizations was seen to increase with a time constant of about 1 min. while pressure relaxed with about the same time constant. These out of equilibrium processes are not easy to analyze. From B to C we certainly produced a large amount of strongly underpolarized solid. The lattice was quite cold since the melting pressure was quite high, and close to the ordering pressure in zero field. When the compression was stopped this cold solid tended to acquire its equilibrium magnetization at that temperature and field. Some of the magnetization was actually recovered and the heat of magnetization released caused the decrease of pressure.

The decrease in magnetization at point B could be related to some warming of the existing magnetized solid due to mechanical friction, but this possibility seems quite remote since only about 20% solid was formed until point B, and compression could be continued until nearly 100% solidification, when a permanent decrease of the NMR signal was observed. Unfortunately we do not have a better explanation at present. This decrease in the total NMR signal was not observed in the subsequent experiments done at Grenoble in a higher magnetic field.

4.2 The Grenoble Pomeranchuk experiment

a) The cell - To obtain some quantitative information on the behavior of solid ^3He in high magnetic fields, as well as higher liquid polarization we made a larger cell, to be precooled by our dilution refrigerator below 3 mK in a 7.2 T field.

This cell had a flexible membrane of 13.6 mm diameter made of 6 layers of Kapton to a total thickness of 94 μm and 24 mm length.

After gluing to the epoxy end pieces the free length of the membrane was 16.1 mm. The liquid ^3He space was further reduced to 14.1 mm length, for a total liquid volume of 2.05 cm^3 .

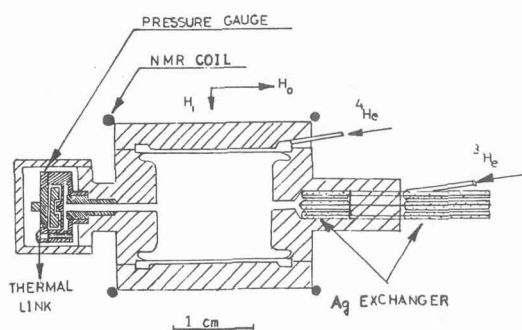


Fig. 8

Fig.8 shows a cut view of the cell.

On one end we glued a Beryllium-Cooper pressure gauge of standard design [29], encased in a vacuum container to keep the reference pressure constant. The connecting channel was 1 cm long and 1 mm in diameter. This channel was expected to be blocked by solid at some stage of the compression (which actually happened at a solid fraction of about 40%) but we did not consider this to be a serious drawback. In fact we expected to solidify most of the liquid at almost constant pressure because of the expected very low initial liquid temperature. The cell also had a volume gauge obtained by evaporating two Ag electrodes on the plastic membrane.

These electrodes were only 4mm wide and were placed in opposite directions in such a way that the RF. field of the NMR coil would be parallel to them. The other pair of electrodes was evaporated inside the ^4He plastic container.

Opposite the pressure gauge we glued the heat exchanger of similar construction to that of the previous cell, with the difference that 3 such exchangers were placed in parallel and that 0.15g

of 400 \AA Ag powder were used with a total area of 0.3 m^2 .

The cell was also provided with a heater, a carbon thermometer (Matsushita 68Ω 1/8 W, sliced to 0.5 mm thickness), and a glass thermometer.

b) The NMR circuit - The NMR detection was done by means of a conventional Q-meter type circuit shown in fig. 9

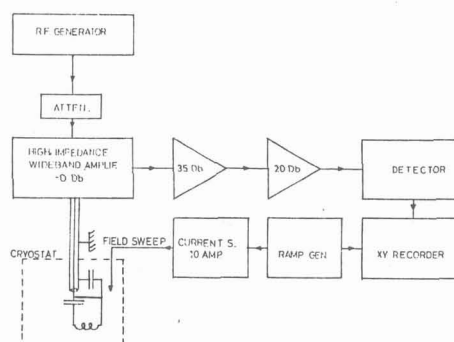


FIG 9 NMR CIRCUIT

The signal area was measured and the height compared to a calibrated 1% change of the RF. level, to correct for drifts of the amplifiers. The NMR coil was of the saddle type, 25 mm in length, 21 mm in diameter, the RF. field being perpendicular to the cell axis. It was tuned to 231 MHz by means of a 0.6 pF capacitor in series and a larger capacitor in parallel to match the circuit as well as possible to 50Ω .

c) Thermometry - The cell was provided with a thermometer made by evaporating two Ag electrodes to a thin (0.16 mm) glass plate $9 \times 11 \text{ mm}$ in size, cut from a microscope slide cover [30]. The very low temperature dielectric behaviour of amorphous materials is now relatively well understood and its temperature variation was shown to be useful as a magnetic field independent thermometer. [31] [32] Commercially available capacitance thermometers [33] could also be used but there are no reports on their usefulness below 10 mK , which can

be limited if the losses and internal thermal resistance are large.

Inside the mixing chamber we had another capacitance thermometer (34) and a Pt NMR coil tuned to 500 KHz.

The mixing chamber capacitance thermometer was calibrated against the nuclear resonance of platinum both C.W. and pulsed. The cell glass thermometer was checked to be practically independent of the ^3He pressure at fixed temperature. A very small effect, linear in pressure, could have been subtracted if necessary. Some calibration points were done in zero field as a function of temperature deduced from the melting curve (28). Unfortunately during a decompression done to adjust the internal pressure of the cell an electric contact of the thermometer broke and we lost our field independent thermometry.

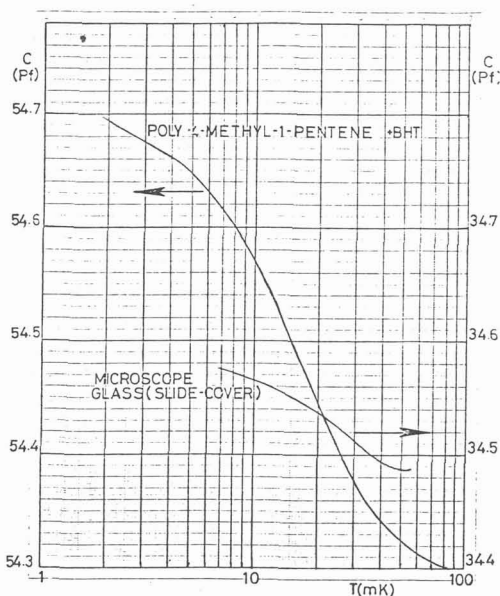


Fig.10 shows the low temperature part of the calibration curves of both thermometers.

The mixing chamber thermometer had a resolution of 30 μK at 2.4 mK when measured with a General Radio 1615 capacitance bridge using 0.5 V at 3430 Hz.

c) Experimental results - Besides the broken thermometer the cell had another problem. The cooling down time constant was 10 times larger than the 3 hours calculated. This was later found to be due to Stycast having leaked and plugged most of the heat exchanger. Since at 7.2 T the melting curve was unknown we made preliminary measurements of it (Fig.2) by allowing the cell, which was directly immersed in the mixing chamber to be in thermal equilibrium with it.

The estimated error at 4 mK was ± 0.5 mK, decreasing with increasing temperature. From the slope of the melting curve we derived the solid entropy at 7.2 T using the Clausius Capeyron equation. This curve is similar to the free spin entropy curve shown in dotted lines, the excess in entropy which appears below 7 mK being due to the antiferromagnetic nature of the exchange interactions. The best Physical model to explain the solid ^3He experiments used 3 and 4 spin exchange, and was reported by M.Roger (20) and J.M.Delrieu (35).

By assuming that the solid was ordered when entropy had fallen to a value so low that cooling ended as a consequence of equilibrium between the heat absorbed and the heat leaks (during very slow reversible compressions) the magnetic phase diagram from 1.0 T to 7.2 T was derived. Entropy measurements at 2.75 mK (T_A) showed that the entropy had fallen quite below that of a paramagnet with antiferromagnetic interactions indicating magnetic ordering. The width of the transition being rather large confirms that the transition is of 2^{nd} order.

These results as well as measurements of the pressure splitting of the A superfluid transition and the linearity of the $P_0 \times H$ diagram, where P_0 is the ordering pressure, are reported separately (11), (12).

Magnetization measurements were done by fixing the dilution refrigerator temperature at some convenient value and allowing the cell to thermally equilibrate. Fixed amounts of solid were then formed (typically 5%) and the magnetization deduced by measuring the areas of the liquid and the solid signals. The liquid signal was corrected from the mixing chamber contribution. The solid fraction for this cell was given by the expression

$$X_s = 2.89 \{ \Delta P_4 - 4.23 \Delta P_3 \}$$

and a maximum of 75% liquid could be solidified.

The liquid magnetization at $T = 0$ was calculated in terms of the saturation magnetization

$$m_{\text{sat}} \text{ by } \frac{m_l}{m_{\text{sat}}} = \frac{\mu H}{k_B T_F^*} \text{ with}$$

$\frac{\mu}{k_B} = 0.8 \text{ mK/Tesla}$ and $T_F^* = 177 \text{ mK}$ at 3.43 MPa (1) which gives $m_l = 0.032 m_{\text{sat}}$.

The NMR signals were checked to be proportional to the solid fractions.

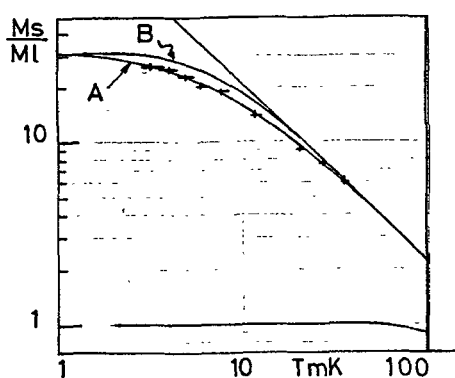


Fig. 11—Solid to liquid magnetization ratio

Experimental points compared to:

a) Mean field Brillouin law with $\theta = 3 \text{ mK}$

b) Brillouin law for non interacting spins

Fig. 11 shows the experimental magnetization points of solid compared to the liquid magnetization taken equal to 1 at $T = 0$. A Brillouin law for an antiferromagnet with $\theta = 3 \text{ mK}$ is seen to fit the points quite well. Also shown for comparison is the Brillouin law for solid ^3He supposed paramagnetic.

The maximum solid polarization obtained was $78 \% \pm 6\%$, the error being due mostly to the mixing chamber contribution to the liquid signal. This value agrees, within the error bars with the one deduced from the $P \times H$ curve slope at the ordering temperature, as analyzed by Roger, Delrieu and Hetherington (20). From this model and the experimental results mentioned above, 14 T would be needed to achieve nearly 100% polarization.

To conclude on the experiments with solid ^3He we would like to comment on a problem related to the growth of magnetization and the features shown in fig. 7. This was observed during solidification experiments. In order to measure the magnetization we formed successive solid fractions at constant temperature. The magnetization per unit solid should have been constant. On the contrary what we saw was that at the lower temperatures after a small initial solid fraction, the new solid was less magnetized. Signals reduced by as much as a factor 3 were observed, if we tried to increase the compression rate (pressure would also increase). If the compression was halted the pressure would decrease, just like in fig. 7 corresponding to heat being released while magnetization was recovering. Compression rates which were very slow compared to zero field compression rates, had to be further decreased and were typically about 5% per hour.

Because of irreversible heat generated when the magnetization of the solid was lower than the equilibrium magnetization the cooling power due to the Pomeranchuk effect was strongly reduced (low compression rates mean low cooling power, which can be rapidly balanced by the magnetic heat due to irreversible heating). Because of the decrease in solid entropy and the onset of order the minimum

temperature at 7.2 T was probably limited to 2.5 - 3 mK.

An experiment we have done using this possibility of making strongly underpolarized solid by fast compression might clarify somewhat this subject.

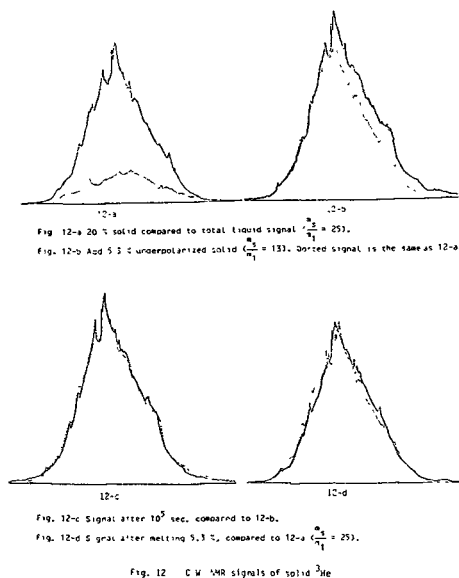


Fig. 12 C W NMR signals of solid ^3He

Fig. 12-a shows the NMR signal due to the formation of 20% solid at the very slow rate of 5.3 % per hour at a temperature around 4 mK. The liquid signal prior to compression, including the mixing chamber contribution is also shown. The magnetization was measured to be 25 times that of the liquid. Fig. 12-b shows the signal obtained after a further quick compression that added 5.3% extra solid, at a rate of 5% per minute. The magnetization of this extra layer was only 13 times that of the liquid, hence this solid was only half magnetized. The compression was then stopped and the cell left for $\sim 10^5$ seconds with the mixing chamber at 3 mK. Fig. 12-c shows the final signal compared to that of Fig. 12-b. It is seen to be nearly identical. Exactly the same amount was then melted in about 10 minutes. The signal after decompression is shown in fig. 12-d, compared to

the initial signal. Apart from small shifts of some of the spikes, the area is the same hence the magnetization did not change during the $\sim 10^5$ seconds.

It seems clear that there was no diffusion of magnetization between the highly magnetized and the lowly magnetized layer.

Knowing the area of the cell and supposing the signal grows uniformly along the cell (the solid signal seems to grow with the same distribution as the liquid signal) we can estimate the spin diffusion coefficient between the two regions. For our cell the wall area was 9 cm^2 and the volume 2.05 cm^3 . 20% solid would correspond to a uniform distribution of about 0.4 mm thickness, and an extra 5% would form a layer of about 0.1 mm. We can derive

$$D \approx \frac{A^2}{\tau} \approx \frac{(2.5 \times 10^{-2})^2}{10^5} \approx 6 \times 10^{-9} \text{ cm}^2/\text{s}$$

This value is quite low but follows the tendency already observed by Johnson, Symko and Wheatley of a sharp decrease of D at large solid polarizations, not yet explained. (36)

If the uniform distribution hypothesis is wrong this value could be larger, but on the other hand after ~ 30 hours no significant change was seen which could mean that D is even smaller.

A short time constant of about 300 sec. was always observed just after growing or melting of solid which correspond to a very superficial relaxation probably at the liquid to solid interface, where many vacancies should exist. This fast relaxation must be responsible for the heat of magnetization release and corresponding decrease of pressure (which adjusts itself to the liquid-solid equilibrium)

The large magnetization of the solid has to be given either by the solid walls of the cell or by

the liquid itself which would lose magnetization to the solid and recover it from the walls because of the fast diffusion of ^3He at those temperatures. If the walls are covered by a fine layer of solid and if we believe the experiment just shown, magnetization can no longer be transferred from the walls to the liquid then to the new solid; the solid layer acts as a magnetic insulator. The only possibility for the solid to be magnetized is via the intrinsic dipole-dipole relaxation of the liquid, intrinsic T_1 could be many thousand seconds which would explain the very slow compression rates needed.

A lack of magnetization is seen as a higher melting pressure at a given temperature. If for some reason T_1 could suddenly be decreased, for a given compression rate magnetization would be less out of equilibrium and the pressure would drop back to a lower value corresponding to the sudden increase of solid magnetization at the interface.

This "backstep" in pressure was observed by Shubert et al (27) to appear after the superfluid A_1 transition. This was first interpreted as a transition in solid but it seems to us that the explanation of Yu and Anderson and of Delrieu (37) agrees better with our observations.

After the superfluid transition, since the temperature must become uniform throughout the liquid, the solid at the top of the cell should migrate to the bottom where the interface would be at a constant pressure (hence temperature).

This would expose the wall, and the supercurrents (or simple diffusion to the wall, which should be quite fast) would then allow fast growing of magnetization. This shows the interest of precooling the cell to the superfluid transition.

An interesting problem concerning polarized liquid ^3He is related to its melting curve. In finite fields the melting curve is depressed (as compared to zero field) because of the reduction of solid spin entropy. The depression ΔP is given, in terms of the liquid and the solid magnetizations, by

$$\Delta P = \frac{1}{v_l v_s} \int_0^H (m_l - m_s) dH$$

where v_l and v_s are the molar volumes of the liquid and solid ^3He . For normal fields m_l is negligible and m_s dominates. (Fig.2). For very high fields ($H \sim T_F$), m_s is saturated and the melting curve will depend on m_l . The extrapolation of m_l to very large polarizations depends on whether the liquid is quasi solid or nearly ferromagnetic.

In the first case a depression of the melting curve of about 0.7 MPa was calculated (5), with liquid phase existing down to $T = 0$. On the other hand, if the nearly ferromagnetic liquid picture is correct, the internal field corresponding to 100% polarization has to be much larger and could lead to the appearance of a triple point, or two triple points, if polarization is incomplete. (5) We see the importance of this kind of measurement. The effect should be relatively small around 300 mK but should increase with decreasing temperature. If the initial solid entropy is very low, the final polarized liquid ^3He should be at very low T and the effect quite easy to observe (roughly 0.2 MPa with 70% polarized liquid at 60 mK).

Some compressions starting from about 45% liquid polarization did show a depression of about 0.1 MPa at the temperatures indicated by the carbon resistor (between 60 and 100 mK) but this "depression" was also found at zero field. The reason could be that large temperature inhomogeneities appear when quickly decompressing, maybe due to frictional heating, in regions far from the thermo-

meter. For a 2 cm^3 cell at 60mK the thermal time constant due to the diffusion of quasiparticles is of several minutes which explains the existence of large thermal gradients.

A much smaller cell is now under test oriented primarily towards the study of the polarized liquid phase, using the experience acquired during the two experiments just described.

5. Conclusion - The method of polarizing ^3He suggested by Castaing and Nozières was shown to be a practical possibility. Solid magnetization was observed to stay in the liquid with relaxation times similar to those of the normal liquid, up to 5 minutes at $P < 1\text{ MPa}$ and $T \sim 100\text{--}300\text{ mK}$.

The magnetization, the melting curve and the entropy diagram of solid ^3He at 7.2 T were also measured. Further evidence of a 2nd order phase transition and extension of the magnetic phase diagram (27) to 7.2 T, confirming the 3 spin + 4 spin exchange model of Roger, Delrieu and Hetherington was reported on another paper presented at this conference (12).

As a consequence of these measurements and of the theoretical model it is found that to obtain polarizations close to 1 on solid ^3He at least a 14 T field must be applied at 3 mK.

Further experiments are now being prepared to gather quantitative information about the strongly polarized liquid ^3He phase. In particular we will try to measure the new melting curve as a function of polarization, the thermal diffusivity and a possible increase in temperature of the phase A_1 superfluid transition due to the megagauss internal magnetic fields, associated with these large polarizations. (38)

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