PRODUCTION OF HIGHLY POLARIZED SOLID ^{3}He IN A POMERANCHUK CELL

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Résumé.- La meilleure façon de produire du solide ^{3}He fortement polarisé dans une cellule est de solidifier dans un fort champ magnétique du liquide ^{3}He prérefroidi. Cet article est conçu comme une présentation pédagogique de la méthode de Pomeranchuk (solidification adiabatique de ^{3}He liquide) appliquée à ce but.

Abstract.- Highly polarized solid ^{3}He in bulk is best produced by solidification, in a magnetic field, of precooled liquid ^{3}He. The paper is a tutorial introduction to the application of the Pomeranchuk method (adiabatic solidification of liquid ^{3}He) for this purpose.

1. Introduction. - The thermodynamic method for spin polarization of liquid ^{3}He, i.e. rapid melting of pre-polarized solid, has caused renewed interest in the question of how to polarize solid ^{3}He. To polarize liquid ^{3}He this way was suggested by Castaing and Nozieres/1/ and later successfully carried out by two experimental groups, Schumacher et al./2/ and Chapellier et al./3/.

For experimental studies of the properties of polarized liquid ^{3}He it is of utmost importance to establish a well-defined and strong initial polarization. At present it appears that the only really efficient method to produce strongly polarized solid ^{3}He is the adiabatic solidification of liquid ^{3}He, also called Pomeranchuk-cooling, in a magnetic field.

It turns out that one is then dealing with solid ^{3}He at so low temperatures, that it no longer behaves like an ideal paramagnet of free nuclear spins. At 1 mK, Halperin et al./4/ found a first order transition to an ordered state, where the magnetization is strongly reduced, according to measurements by Prewitt and Goodkind /5/. In magnetic fields stronger than 0.4 Tesla the transition changes its character and the transition temperature increases (Kummer et al./6/), reaching 3 mK in 7 Tesla (Godfrin et al./7/). Some of these observations have confirmed the existence in solid ^{3}He of spin-spin interactions thousands of times stronger than the dipole interactions, as was previously inferred from data at higher temperatures.

The aim of the present paper is to describe the Pomeranchuk method and to discuss the behaviour of an experimental cell in sufficient detail to enable the reader to evaluate Pomeranchuk cell data with a sound scepticism. The limitations on polarization possibly imposed by the phase transition mentioned above, the actual observation of polarized liquid, and problems with relaxation phenomena in the spin system, are treated in the companion articles by M. Chapellier/MC/ and by G. Frossati /GF/.

2. Entropies of Liquid and Solid ^3He. Heat Transfer.

If we treat solid ^3He as an ideal paramagnet, with independent spins, then a polarization around, say, 80% is obtained by a ratio of magnetic field to temperature, B/T of 1.3 Tesla/mK. In a 7 Tesla magnet the sample must be cooled to 5 mK, a temperature attainable with dilution refrigerators. The entropy reduction needed is 0.5 R ℓn 2, i.e., half the high temperature entropy of the spin system must be removed. According to present knowledge this is an unsurmountable problem, as we shall see now.

Heat must be removed from the sample through its boundaries to other solid materials. As first shown by Kapitza, a heat current $\dot{q}$ between two bodies at low temperature, is accompanied by a temperature jump right at the boundary: $\Delta T = R_K \dot{q}$, where the Kapitza resistance R_K is inversely proportional to the area of contact and depends strongly on temperature.

The Kapitza resistance between solid ^3He and any other material has not been measured at these low temperatures. There are no indications, however, that is should be spectacularly different from the Kapitza resistance for liquid ^3He, which has been measured to some materials down to a few mK. Above 10 mK, R_K depends on temperature as T^{-3} . Between the celebrated heat exchanger material, sintered silver powder, and liquid ^3He, the Kapitza resistance below 10 mK increases only as T^{-1}. Frossati/8/ has found $R_K \cdot \sigma = 1300/T(m^2K^2/W)$. σ is the contact area

Fig.1 shows the entropy of solid ^3He in zero magnetic field (measured, /4/,/9/), and in 7 Tesla (as calculated for a free spin paramagnet), along with the entropy of liquid ^3He

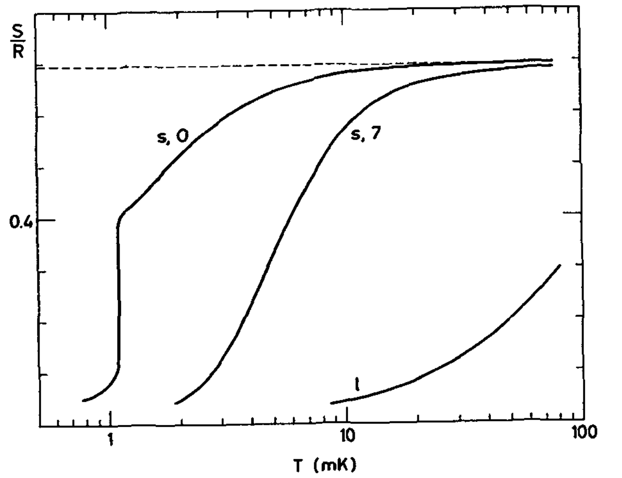

Fig.1. - Molar entropies, divided by the gas constant R , for ^3He. s,0 : solid in zero field (measured /9/). s,7: solid in 7 Tesla (calculated for free spins).ℓ : liquid (measured and extrapolated /9/).

(measured, /9/). It is seen that for the solid, most of the entropy reduction, even in 7 Tesla, has to be done below 10 mK. If we want to refrigerate a 1 cm^3 cube of solid to 5 mK in 7 Tesla and take the above value for the Kapitza resistance at 10 mK, the cool down time is estimated to be of the order 1 year. One obvious way to reduce the time needed for cool-down would be to increase the contact area. With sintered copper or silver powder, the surface-to-volume ratio could be improved rather easily 10^3-10^4 times.

At this point, then, a second limiting factor shows up: the poor thermal conductance of solid ^3He. Again, no direct measurements exist, but thermal time constants are known to be extremely long, already at 20 mK. In the experiment of Prewitt and Goodkind, this difficulty was overcome by having the sample contained in pores of submillimetre size in a sintered copper powder. Having in mind our wish eventually to melt the polarized solid and to preserve spin polarization for the longest time possible we want to limit spin relaxation at surfaces, and this obviously calls for a sample cell with an open space. Relaxation time data are being collected, but the present conviction is that dimensions of the open space should be a centimeter or more /MC/.

Fortunately, nature has provided one more path. The entropy of liquid ^{3}He has fallen to $0.5\ R\ \ell n\ 2$ already at temperatures around 80 mK (Fig.1). (In the magnetic fields concerned here, the liquid entropy is not expected to be field dependent). So, in principle it should be possible by isentropic solidification of the liquid precooled to 80 mK to obtain solid with an entropy corresponding to a polarization of 80%. This argument implies even, that the resulting polarization will be independent of the size of the applied magnetic field.

In a real experiment, one needs to keep heat leaks to the sample to a minimum. Therefore, the walls of the sample chamber should be close to the final temperature T_f determined by B/T_f = 1.3 Tesla/mK for complete solidification to a paramagnetic phase. The thermal conductivity of liquid ^{3}He is relatively high at these temperatures, 0.33/T(mW/m), and it is possible to precool the liquid via a heat exchanger to nearly the temperature of the refrigerator in a reasonable time. As recently demonstrated by Osheroff et al./10/ it is, in fact, possible to grow crystals of solid ^{3}He at 0.5 mK, by following this path.

The Pomeranchuk method is so powerful, that a strong magnetic field may be needed to prevent the solid ^{3}He formed from entering the low temperature antiferromagnetic phase /GF/. Johnson et al./11/ obtained solid polarized to around 50% from liquid precooled to 45 mK, in a field of 5.5 Tesla.

3. Liquid and Solid ^{3}He in a Pomeranchuk Cell.

The solidification or melting pressure P_m of ^{3}He has a minimum of 2.93 MPa near 300 mK. Towards zero temperature P_m increases to 3.44 MPa. A Pomeranchuk cell is a device by which the total volume of a ^{3}He sample can be controlled at these pressures and temperatures.

Fig.2 shows the all-plastic construction developed by Frossati/12/. The cell body is made of epoxy. The volume variation is effectuated by the movement of a thin flexible cylindrical wall consisting of layers of Kapton foil with a total thickness around 0.1 mm for a sample space of 10 mm diameter.

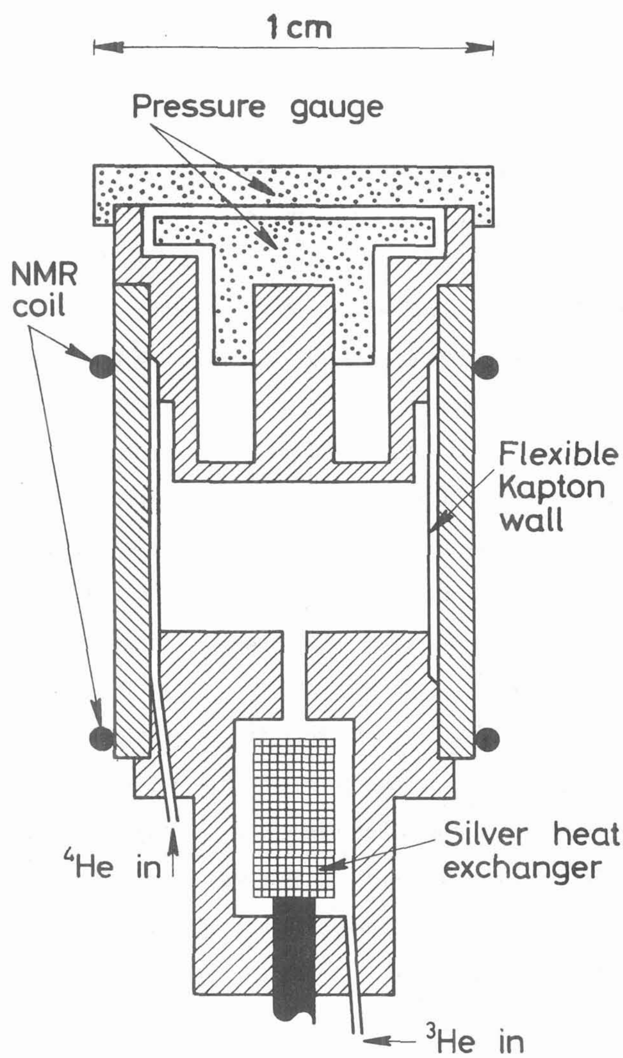

Fig.2. -The plastic Pomeranchuk cell of Frossati/12/. Its operation is described in the text.

The position of the flexible wall is determined by the difference $P_3 - P_4$ between the sample pressure P_3 and the pressure P_4 of liquid ^{4}He injected on the outside of the wall. The position is influenced by absolute pressure also, depending on the stiffness of the cell body. ^{4}He becomes solid around 2.6 MPa so that elastic tension of the Kapton wall must supply the remaining pressure difference up to $P_3 \sim 3.44$ MPa. Yet, the wall must be flexible enough to yield a volume change around 8%, as P_4 varies between 0 and 2.6 MPa. Total solidification of an all liquid ^{3}He sample calls for a 5% change. If at the same time P_3 goes from 2.93 to 3.44 MPa the high compressibility of liquid and of solid ^{3}He, ca. 5×10^{-2} MPa^{-1}, gives rise to an additional volume change of 2.8%.

When P_3 is above 2.93 MPa, the capillary connecting the cell with room temperature containers and manometers will be blocked with solid ^{3}He in a section of tubing with temperatures around 300 mK. The position x of the liquid-solid interface in the tube is determined by the temperature distribution $T(x)$ and P_3 by $P_m(T(x)) = P_3$. Because of this solid block, P_3 has to be measured by a capacitive pressure gauge in the cell itself. The type shown in Fig.2 has the advantage, that its connection with the sample volume cannot be plugged with solid ^{3}He. On the other hand, its readings are found to be influenced rather strongly by P_4.

For small pressure changes the change in volume is proportional to the change in pressure difference:

$$\Delta V = k\Delta(P_3 - P_4) . \qquad (1)$$

k plays the role of an elastic constant of the flexible wall. Volume changes may also be detected very sensitively by a capacitance gauge consisting of two metal films facing each other across the ^{4}He space.

Other Pomeranchuk cells have relied on flexible metal parts for the volume variation: a system of bellows /10/, a thin-walled metal tube / 6 / or a thin flexible diaphragm forming one side of a flat, lense-shaped sample volume /9/. The design in Fig.2 has the advantage of a compact cylindrical shape inside plastic walls so that an NMR coil detecting signals from the whole sample may be placed around it. Its disadvantages are that pressure gauge readings do not reproduce well and depend on P_4, and that the cell is rather fragile and may be destroyed by a negative P_3-P_4.

As long as the fill line is plugged, a constant number n_3 of moles of ^{3}He stays enclosed in the cell. If pressure P_3 and volume V are accurately known the amounts n_s and n_ℓ of solid and liquid, respectively, may be found from:

$$n_3 = n_s + n_\ell \qquad (2)$$

$$V = n_s v_s + n_\ell v_\ell \qquad (3)$$

where v_s and v_ℓ are the molar volumes at that pressure. Pressure needs to be specified because of the high compressibility. Quite often, e.g. in relative measurements of spin polarization, it is sufficient to know the <u>fractions</u> of liquid and solid, x_ℓ and x_s. These may be determined from measurements of relative volume changes.

Corresponding to changes in temperature ΔT, in pressure, ΔP_3 and in the amount of solid Δn_s, there is a relative change in volume determined by

$$\Delta V/V = \bar{\alpha}\Delta T + \bar{\kappa}\Delta P_3 + (v_s - v_\ell)\bar{v}^{-1}\Delta x_s \qquad (4)$$

$\bar{\alpha}$, $\bar{\kappa}$ and $\bar{V}$ are averages of the thermal expansion coefficients, compressibilities and molar volumes of solid and liquid, weighted in proportion to their volume or molar fraction as appropriate. Some experimental values are listed in /9/, but for orders of magnitude we may note that heating the liquid from 10 to 50 mK at constant pressure produces a (negative) volume change corresponding to solidification of 0.3% of the sample at constant temperature. Thermal expansion of solid would be one fifth of this. The compressibility term may be considerably larger. If the heating above were done at the melting pressure the change in P_m would produce a $\Delta V/V$ simulating melting of 14% of solid at constant pressure.

A Pomeranchuk compression would usually start with a fully liquid sample precooled to the lowest possible temperature. Reducing the sample volume at a constant rate $\dot{V}$ and at constant temperature, P_3 will be observed to increase at a rate $\dot{P}_3$ determined by $\dot{V}$ and κ_s through eq.(4). When P_3 reaches P_m at the sample temperature, solidification starts, and there is a decrease in $\dot{P}_3$ - the sample effectively becomes "softer", because of the added degree of freedom. Measuring $\Delta V/V$ and ΔP_3 from this clear signature in the trace of P_3 versus time, $x_s = \Delta x_s$ may be calculated from (4).

The spin polarization, or magnetization, is measured by the area under the NMR resonance line at any time in proportion to the all-liquid line. Liquid susceptibilities for $T \to 0$ are tabulated in /13/. Since the liquid susceptibility is independent of T at the low temperatures concerned here, any additional polarization is in the solid. Dividing the solid magnetization contribution with x_s one finds the average polarization of the solid.

A word of caution is in place, however. Several observations /9/, /3/ indicate that solid ^{3}He formed during a compressional cooling experiment does not follow the cell temperature immediately. As a consequence, the temperature of the solid formed has a spatial variation. The relaxation rate of this temperature distribution will depend on the actual temperatures, on the amount and distribution of solid, and on the magnetic field. Relaxation times may be thousands of seconds. So, at the end of a compression in a magnetic field, we may expect the local spin polarization to vary with position inside the solid. Consequently, spin polarizations calculated by the recipe above represent lower bounds only on the polarizations obtained.

Without special and careful precautions /10/ solid grows irregularly in the Pomeranchuk cell. Establishing a well specified (if not homogeneous) distribution of spin polarization will be a conditio sine qua non for progress in studies of polarized liquid ^{3}He.

Actual results and some NMR aspects are discussed by Frossati /GF/ and Chapellier /MC/.

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