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## Nuclear polarization through optical pumping of gaseous $^3\text{He}$ below 1 K

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**Résumé.** — La méthode du pompage optique qui permet d'orienter nucléairement  $^3\text{He}$  gazeux a pu être étendue au domaine des températures en dessous de 1 K. L'utilisation d'un enduit superfluide de  $^4\text{He}$  couvrant la paroi interne de la cellule a permis de réduire très fortement la relaxation nucléaire intense qui serait produite par du pyrex nu. On obtient ainsi des temps de relaxation nucléaires relativement longs ( $T_1 \geq 10^3$  s), même aux températures les plus basses accessibles dans l'expérience ( $T \cong 500$  mK).

**Abstract.** — We have extended to temperatures below 1 K the optical pumping technique for polarizing the nuclei of ground state  $^3\text{He}$  atoms in the gas phase. A superfluid  $^4\text{He}$  film covering the surface of the pyrex cell protected the  $^3\text{He}$  atoms from the rapid depolarization processes that would be experienced in the presence of bare pyrex walls. Relatively long relaxation times of order  $10^3$  s were observed at temperatures down to 500 mK, the lowest temperature accessible with the present apparatus.

There are several motivations for doing optical pumping experiments in gaseous  $^3\text{He}$  at the lowest possible temperatures. First, theory predicts [1] that, at very low temperatures, gaseous spin polarized  $^3\text{He}$  ( $^3\text{He} \uparrow$ ) should exhibit several unusual macroscopic quantum properties such as enhanced viscosity or heat conduction, and spin waves [2]. Optical pumping may provide the necessary nuclear polarization in attempts to observe these low temperature effects. There are similarities between  $^3\text{He} \uparrow$  and spin polarized hydrogen ( $\text{H} \uparrow$ ) for which spectacular macroscopic quantum effects — in particular a Bose-Einstein condensation — have also been theoretically predicted [1]. More generally, even in the absence of high nuclear polarization, the properties of gaseous  $^3\text{He}$  or  $\text{H} \uparrow$  are an interesting subject in atomic physics, since these systems are the only gases that can be studied at temperatures below 1 K (all other systems liquefy or solidify). Another motivation to do optical pumping experiments at low temperatures is related to surface physics : it is well known [3] that optical pumping experiments can be aimed at surface physics studies through measurements of relaxation probabilities due to atom-wall interactions.

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Previous experiments [5, 6] with optically pumped  $^3\text{He}$  have shown that low temperature techniques can provide reproducible and rather well controlled wall surfaces. These experiments show in particular that the crucial number which governs the wall relaxation of nuclear spins at low temperatures is the adsorption energy  $\Delta W$  of an atom on the walls. Since the bulk relaxation times in dilute gaseous  $^3\text{He}$  are very long (of the order of  $10^5$  h [7-9]), nearly all the spin depolarization occurs while the atoms are adsorbed on the inner surface of the container and experience the effect of local magnetic fields created by paramagnetic impurities. Under these conditions, the rate of relaxation of the whole sample obeys :

$$\frac{1}{T_r} \propto \exp\left[q \frac{\Delta W}{kT}\right], \quad (1)$$

where  $q$  is a number of the order of 1 <sup>(1)</sup>. When  $kT \ll \Delta W$ , the exponential function has a very fast temperature variation, and it is found [6] in practice that the optical pumping process is completely quenched by the wall relaxation whenever  $kT \lesssim q \Delta W/15$ . Consequently, only walls with  $^3\text{He}$  adsorption energies below approximately 10 K can be used for optical pumping below 1 K.

Since the adsorption energy of one  $^3\text{He}$  atom on a glass surface is of the order of 200 K [10], uncoated glass walls are clearly completely inappropriate. An efficient way to reduce  $\Delta W$  is to coat the walls of the cell with several layers of solid  $\text{H}_2$ . This technique makes it possible to do optical pumping at  $T = 4.2$  K [5]; nevertheless,  $q \Delta W$  is still of the order of 30 K [6] and therefore too large for experiments below 1 K. The last remaining possibility (as far as we can see) is then to use walls coated with liquid  $^4\text{He}$ ; the larger mass of the  $^4\text{He}$  atoms, and consequently their smaller zero point energy, allows them to condense and form Van der Waals films on surfaces at higher temperatures than the  $^3\text{He}$  atoms, so that most of the latter remain in the gas phase. The binding energy of  $^3\text{He}$  atom on the surface of superfluid  $^4\text{He}$  is  $\Delta W \cong 5$  K [11], which is low enough for the required purpose. In addition, superfluid  $^4\text{He}$  films tend to cover very effectively all surfaces available to them, due to their high mobility. They have already been used with great success for the conservation of  $\text{H} \uparrow$  down to temperatures below 100 mK [12] (the corresponding adsorption energy being only of the order of 1 K [12, 13]). The use of superfluid  $^4\text{He}$  coatings seems therefore very attractive.

Actually, a more careful investigation shows that some problems might arise from the use of these coatings in experiments dealing with  $^3\text{He}$  instead of  $\text{H} \uparrow$  atoms. The former can dissolve inside superfluid  $^4\text{He}$ , where their binding energy with respect to free space is approximately 2.8 K [11]; inside the liquid, they behave like free particles (with an effective mass about 2.5 times the normal mass [14]). If some of the  $^3\text{He}$  atoms penetrate inside the superfluid film and can reach the glass walls, it is not obvious that the presence of the film will reduce the nuclear relaxation rate at all. Fortunately, it seems now well established that the  $^4\text{He}$  atoms tend to exclude the  $^3\text{He}$  from the first layer (and possibly the second) covering a glass surface [15]. This differential adsorption effect between the two isotopes is again a consequence of the zero point motion; it presumably explains why previous experiments [16, 17] on liquid  $^3\text{He}$  confined on grafoil have shown that the longitudinal relaxation time of the nuclei can be significantly enhanced by the presence of a few  $^4\text{He}$  layers. Similar effects should occur in superfluid Van der Waals films of  $^4\text{He}$  on pyrex glass and prevent the  $^3\text{He}$  atoms from coming close to the walls. Encouraged by these prospects, we decided to test the efficiency of superfluid  $^4\text{He}$  coatings for  $^3\text{He}$  optical pumping experiments.

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<sup>(1)</sup>  $q = 2$  or  $1$  depending on whether the correlation time of the magnetic perturbation on the nuclear spins is equal to or shorter than the adsorption time of the atoms on the surface.

### 1. Experimental arrangement.

A schematic diagram of the experiment is shown in figure 1. The  $^3\text{He}$  atoms are contained in a pyrex cell C (3 cm in diameter) which is cooled down by a  $^3\text{He}$  refrigerator R in the temperature range 0.5 K-1 K. A superfluid helium bath  $B_t$  provides an efficient thermalization between the cell and the heat exchanger E. The temperature is deduced from the measurement of the  $^3\text{He}$  vapour pressure just above the liquid level through the tube  $l_p$ .

In addition to the  $^3\text{He}$  atoms ( $^3\text{He}$  gas filling pressure : 1 torr)<sup>(2)</sup>, the cell C contains the coating materials : 1.5 torr of hydrogen, which condenses first as the cell is cooled and provides a preliminary coating<sup>(3)</sup> of the glass (« undercoating »), as well as 2 torr of  $^4\text{He}$  gas, which, at low temperatures, forms the superfluid coating itself. From the bulk vapour pressure, one would predict that the  $^4\text{He}$  gas starts to condense at  $T_0 = 0.8$  K, is half condensed at 0.7 K, and that 99.5% of it is condensed at 0.5 K. In fact, the first helium layers are certainly condensed at a temperature higher than  $T_0$ , due to the Van der Waals attraction of the helium atoms by solid hydrogen. If the inside surface of the cell were perfectly smooth, with an area equal to the geo-

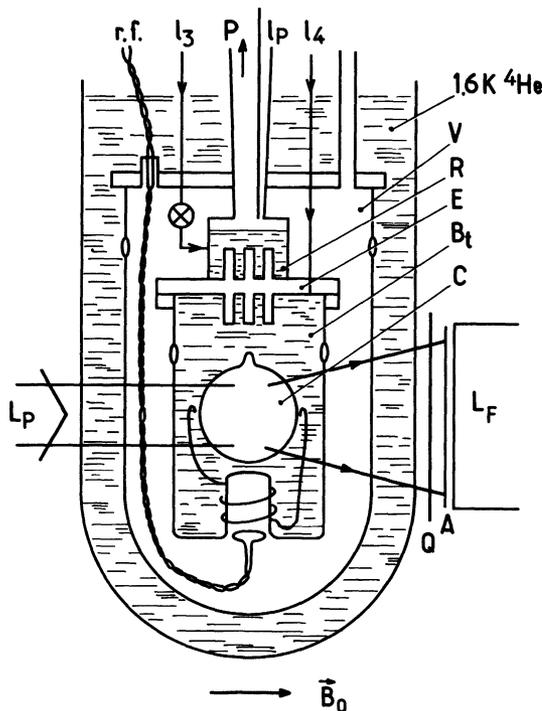


Fig. 1. — Schematic diagram of the apparatus. a) cryogenics :  $B_t$  : thermalization  $^4\text{He}$  bath; C : pyrex cell containing the optically pumped  $^3\text{He}$  atoms; E : copper heat exchanger;  $l_3$  : liquid  $^3\text{He}$  injection line;  $l_4$  : filling tube for  $B_t$ ;  $l_p$  : vapour pressure measurement tube; P :  $^3\text{He}$  pumping line; R :  $^3\text{He}$  refrigerator; V : vacuum pumping line. b) optics : A : analyzer;  $B_0$  : 7 G magnetic field;  $L_F$  : discharge fluorescence light;  $L_p$  : laser beam providing the  $1.08 \mu$  pumping light; Q : rotating quarter wave plate; RF : 10 MHz radiofrequency producing a weak discharge in the cell.

<sup>(2)</sup> All quoted pressures refer to filling pressures at room temperature of the sealed cells.

<sup>(3)</sup> The hydrogen undercoating has been introduced as a supplementary precaution to increase the distance of the  $^3\text{He}$  atoms from the glass.

metrical one, the maximum  $^4\text{He}$  film thickness would be 130 Å. Actually, there are good indications from experiments with  $\text{H}_2$  coatings at higher temperatures [6] that the area is approximately twice the geometrical one. It is difficult to know how this will affect the  $^4\text{He}$  film properties, since the scale of the roughness of the glass surface is not known, and since the surface area can be changed by the presence of the solid hydrogen film. For all these reasons, the figures given above for  $T_0$  and the film thickness are uncertain by a factor of the order of two. As for  $^3\text{He}$ , it has a much higher vapour pressure than  $^4\text{He}$  and remains gaseous down to 0.28 K.

The nuclear orientation of the  $^3\text{He}$  atoms is achieved by the standard optical pumping technique [18] : metastable  $2^3\text{S}_1$  helium atoms are produced by a weak R. F. discharge at 10 MHz ; the power dissipated in the cell is less than 10 mW, and can be absorbed by the  $^3\text{He}$  refrigerator in the temperature range 0.6-1 K. The metastable atoms are optically pumped by a circularly polarized laser beam provided by the colour centre single mode laser described in reference [19]. The typical pumping power was 10-20 mW. The laser frequency was tuned to the strongest of the two  $2^3\text{S}_1, F = 3/2 \rightarrow 2^3\text{P}, F = 3/2$  transitions (called  $\text{C}_5$  in [20]), which was found to be the most efficient component for optical pumping at very low temperatures. The orientation is transferred to the ground state by collisions (metastability exchange collisions, deexcitation on the wall, metastable-metastable collisions, ... ; the exact weight of each of these processes at low temperatures is not known). This indirect pumping cycle leads to long pumping times, of the order of 100 s. A pair of Helmholtz coils produces a static homogeneous magnetic field  $B_0$  (7 G ; nuclear magnetic Larmor frequency 20 kHz) in order to avoid the disorientation of the spins by magnetic gradients due to the environment (<sup>4</sup>). The nuclear polarization is monitored by measuring the degree of polarization  $\mathcal{P}$  of the visible light  $L_F$  emitted by the  $3^1\text{D}$  and  $3^3\text{D}$  excited atoms in the discharge [21]. The polarization of this light is analyzed by a rotating quarter wave plate Q followed by a static linear analyzer A. The modulation amplitude of the photocurrent is directly proportional to  $\mathcal{P}$ .

## 2. Experimental results.

The build-up of a  $^3\text{He}$  nuclear orientation by optical pumping has been obtained for temperatures between 0.65 K to 0.8 K. The destruction of this orientation by nuclear magnetic resonance is also easily observed. The maximum value of  $\mathcal{P}$  is  $3.5 \times 10^{-3}$ , which corresponds to a 3 % nuclear polarization (<sup>5</sup>) according to the conversion factor of reference 21. The mere fact that optical pumping is effective indicates that the wall relaxation time is longer than 10 s. Actually, the observed signal build-up time is limited by various processes occurring in the discharge, such as excitation, or ionization of the ground state atoms, exchange collisions with ions, ... On the other hand, in the absence of a discharge, the relaxation of the nuclear orientation occurs essentially through atomic collisions on the coated walls (with a small contribution from magnetic inhomogeneities, see above). To measure this wall relaxation time at a given temperature  $T$ , one proceeds as follows : one first polarizes the spins by optical pumping at a temperature  $T' \cong 650$  mK (<sup>6</sup>) and one measures the initial degree of polarization  $\mathcal{P}(0)$ . Then, the discharge is turned off and the temperature quickly adjusted to the desired value  $T$ . After a time delay  $\Delta t$ , the temperature is set again at  $T'$ , the discharge is switched on and one measures the remaining degree of polarization  $\mathcal{P}(\Delta t)$  (Fig. 2). Assuming an exponential decay of the nuclear

(<sup>4</sup>) The relaxation time due to the field inhomogeneities has been measured in 7 G at 4.2 K and is of the order of  $10^4$  s.

(<sup>5</sup>) This value seems to be rather insensitive to an increase of the laser power beyond 20 mW.

(<sup>6</sup>) Lower temperatures cannot be obtained because of the heat dissipated by the discharge ; on the other hand, at higher temperatures, the presence of much  $^4\text{He}$  in the gas phase is not desirable for optical pumping.

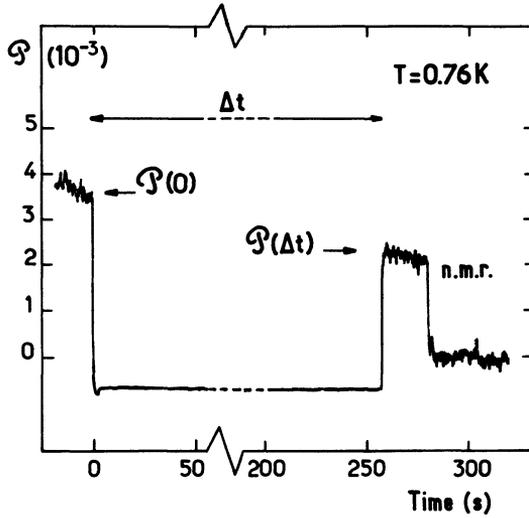


Fig. 2. — Measurement of  $T_r$  : the discharge is turned off during a time interval  $\Delta t$ . The measured quantity,  $\mathcal{P}$ , is the degree of circular polarization of the visible light emitted by the discharge, which is proportional to the nuclear orientation in the ground state. The orientation is finally destroyed by NMR.

orientation, one extracts the relaxation time  $T_r$  from the formula :

$$\frac{\Delta t}{T_r} = \text{Ln} \frac{\mathcal{P}(0)}{\mathcal{P}(\Delta t)}. \tag{2}$$

The signal to noise ratio is typically 15, and the uncertainty in  $T_r$  is quite large, of the order of 20 %. We have measured  $T_r$  for different temperatures between 1 K and 0.5 K. The results are shown in figure 3, where  $T_r$  is plotted on a logarithmic scale against  $1/T$ . A variation of  $T_r$  governed by an adsorption energy (Eq. 1) would then appear as a straight line with a slope  $-\Delta W$ . The temperature  $T_0$  defined above in terms of the  $^4\text{He}$  condensation is indicated by the vertical line.

Clearly, long relaxation times are observed for  $T > T_0$ , indicating that a few layers of  $^4\text{He}$  are already condensed for  $T > T_0$  and act efficiently to prevent relaxation. The shaded region below  $T_0$  corresponds to the formation of the saturated thick film. At the end of this region, half of the  $^4\text{He}$  is theoretically condensed on the wall. This situation corresponds approximately to the maximum value of  $T_r$ . For lower temperatures, and down to 0.5 K, the relaxation time remains long, but slightly decreases. No part of the curve exhibits a slope corresponding to the binding energy of a  $^3\text{He}$  atom either inside the  $^4\text{He}$  film ( $\Delta W = 2.8$  K) or on the surface of the film ( $\Delta W = 5$  K), in the temperature range available in our experiments.

Preliminary measurements on other cells filled with various  $^4\text{He}$  pressures seem to indicate a similar behaviour with different values of  $T_0$ . We have also observed that the hydrogen undercoating is not absolutely necessary : long relaxation times have also been observed with  $^4\text{He}$  only although they are shorter than with hydrogen undercoating. Improved detection of the nuclear orientation using NMR is planned for the near future. We hope to obtain in this way more systematic and better measurements.

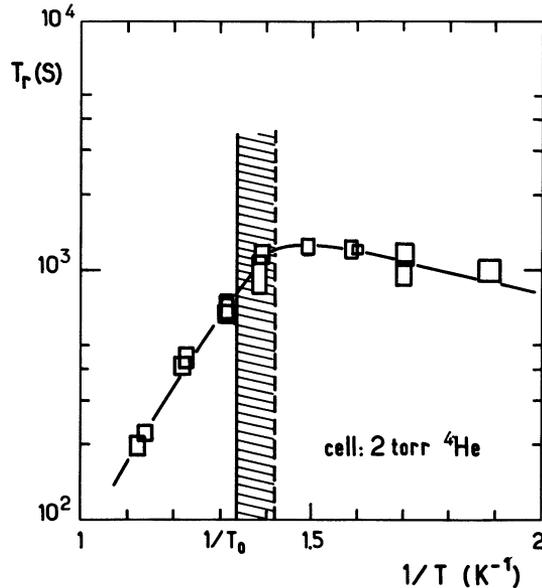


Fig. 3. — Wall relaxation time  $T_r$  on superfluid coating versus the inverse temperature  $1/T$ . The definition of  $T_0$  is given in the text and corresponds qualitatively to the temperature of the  $^4\text{He}$  film formation ; at the end of the gray region, the density of  $^4\text{He}$  in the gas phase has been divided by a factor 2. The solid line is just a guide for the eye.

### 3. Discussion and conclusion.

Our results show that optical pumping of  $^3\text{He}$  works down to 600 mK and that even at the lowest temperature ( $T \cong 500$  mK) reachable with our experimental set up, the nuclear relaxation time can be longer than 10 min. Actually, it seems likely that optical pumping could be performed directly at lower temperatures with a more powerful cooling system able to absorb the power dissipated by the gas discharge, which is necessary for optical pumping. In this respect, the situation differs from that obtained with solid cryogenic coatings such as  $\text{H}_2$ ,  $\text{D}_2$  and  $\text{Ne}$ , where uncontrolled effects of the discharge on the coatings themselves limit the lowest optical pumping temperature to a value where  $T_r$  (relaxation time in the absence of a discharge) is still very long [6]. One possible explanation of this phenomenon relies on the presence of holes, steps, strong binding sites, etc... created by the discharge on the surface of the solid coating ; a liquid surface, especially for a superfluid liquid, is not likely to give rise to such phenomena.

As this stage, no exponential variation of  $T_r$  as a function of the inverse temperature has been found ; a slope of 5 K would correspond to  $^3\text{He}$  atoms sitting on the surface, and a slope of 2.8 K to  $^3\text{He}$  atoms inside the liquid, but both these figures might be different for thin superfluid films [22]. Further experiments are clearly needed to understand the physical origin of the nuclear relaxation. One can nevertheless consider as very encouraging the absence of evidence for a fast exponential variation of  $T_r$  at low temperatures : if one extrapolates our results to lower temperatures,  $T_r$  is found to be a few minutes at a temperature  $T \cong 200$  mK ; in other words, the wall nuclear relaxation apparently does not put any lowest limit on the temperatures accessible to  $^3\text{He}$  optical pumping (at 250 mK, the saturating vapour pressure of  $^3\text{He}$  corresponds to a number density of  $10^{16}$  at./ $\text{cm}^3$ , so that much lower temperatures are of little interest for optical pumping experiments anyway).

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