

Nuclear Polarization and Heat Conduction Changes in Gaseous ^3He .

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Abstract. – Although it is a dilute nondegenerate system, a gas of $^3\text{He} \uparrow$ (spin-polarized ^3He) can exhibit interesting quantum effects due to particle indistinguishability. One of these effects is a variation of the heat conduction coefficient as a function of the nuclear polarization. An experiment is described where this change has been observed and measured. The nuclear polarization is obtained by optical pumping at a wave-length $\lambda = 1.08 \mu\text{m}$ with a LNA crystal laser. The experimental results are in reasonable agreement with theoretical predictions, but the discrepancies are sufficiently clear to suggest that calculations should be slightly revised (including, possibly, a new fit of the He-He interatomic potential).

It is well known that the helium liquids at low temperatures show many interesting quantum properties related to the statistics of the particles (Fermi for ^3He , Bose for ^4He) [1]. The density of these liquids is relatively high ($n \geq 10^{22}$ atoms per cm^3) and these systems are degenerate at the temperatures where their quantum properties are observable. In the present article, we study a quantum effect, related to statistics, which occurs in a dilute gas: the density of our samples is approximately 10^4 times smaller than in a liquid and the system is clearly nondegenerate. Moreover, the quantum effect studied is actually independent of density; it consists in a change of the heat conductivity of the gas when the nuclear spins are polarized. Such a change is, to some extent, paradoxical, since the nuclear spins are so weakly interacting with any other variable of the system. The effect can only be understood quantum mechanically: roughly speaking, in a fully polarized sample of ^3He ($^3\text{He} \uparrow$), all atoms are completely indistinguishable fermions (the direction of their

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spins cannot be used to distinguish one atom from another), which have a totally antisymmetric orbital wave function; two atoms cannot be at the same point of space and they have a small probability to be found at a distance less than λ_T (the de Broglie wavelength). Then, if

$$\lambda_T \gg a_0 \quad (1)$$

(where a_0 is the interaction potential range), the Pauli principle forbids atoms to come close enough to have significant interactions: they propagate almost like free atoms. In other words, the scattering cross-section is strongly reduced. Consequently, the mean free path is larger in $^3\text{He} \uparrow$ than in ordinary ^3He , where collisions between opposite spin states are not affected by the Pauli principle. This implies, in turn, similar variations for the heat conductivity [2]. The same type of effect occurs in dilute ^3He - ^4He solutions [3]. In a ^3He gas, precise theoretical predictions have been given by Lhuillier [4].

Other quantum effects occur in polarized gases when condition (1) is fulfilled. One example is the existence of spin waves [5, 6], which have been observed recently in various nondegenerate gases [7-9]. Their study requires NMR techniques to measure the orientation of the nuclear spins. In contrast, in the present heat conduction experiment, the detection scheme is «more macroscopic»: it is merely a temperature measurement and it does not involve the spins in any direct way. In this respect, our measurements can be compared to those of ref. [10] (second sound in $^3\text{He} \uparrow$ - ^4He solutions) and [11] (viscosity in liquid $^3\text{He} \uparrow$).

Figure 1 shows a general sketch of the experiment. The nuclear polarization is obtained by optical pumping with a LNA laser which has been recently developed in our laboratory [12] and delivers $\approx (200 \div 300)$ mW at $\lambda = 1.083 \mu\text{m}$. A «double cell» technique is used, as described in more detail in [13]. It consists of one cell at room temperature where a weak discharge is sustained in the gas and nuclear polarization is created by optical pumping and one measurement cell at low temperature. The set of the two pyrex glass cells, connected by a 75 cm long tube, was sealed after initial cleaning and filling with ^3He (pressure: 2.2 Torr) and H_2 (pressure: 0.62 Torr). When the lower cell is cooled, the ^3He pressure drops to ≈ 0.5 Torr and the hydrogen forms a solid coating on the inner cold walls of the container. This coating strongly reduces the nuclear relaxation rate [14]. The lower cell is located inside a nonmagnetic pyrex glass cryostat. A series of large coils is used to create a vertical homogeneous field of 3 G.

Figure 1 also shows an enlargement of the lower cell with more details of its geometry. It is a cylinder with two parallel glass plates, 2 mm thick and 3 cm in diameter, 1 cm apart, externally silvered to ensure a uniform temperature. The glass side walls are ≈ 0.5 mm thick. The temperature θ_1 of the upper plate is fixed by the pumped liquid ^4He bath of the internal cryostat. The lower plate, at temperature θ_2 , faces an evacuated space between the two liquid- ^4He baths (see fig. 1), which can be pumped below 10^{-6} Torr by a diffusion pump. An electrically controlled heating power \dot{Q} can be applied to the bottom plate by manganin resistive wires glued on to its silvered surface. Temperatures θ_1 and θ_2 are monitored with Allen-Bradley resistors R_1 and R_2 (100 Ω at room temperature). Precise measurements of R_1 and R_2 are performed with a resistor bridge (AVS-45 RV-Elektroniikka); R_1 is calibrated in temperature from the pressure of the internal ^4He bath⁽¹⁾; R_2 can actually be

⁽¹⁾ A 5-parameter logarithmic law of the type

$$\log \theta = \sum_{n=0}^4 A_n [\log R]^n$$

is used for a computer fit of the temperature θ to the resistor values R in the whole temperature range (1.6 \div 4.2) K).

simultaneously calibrated assuming $\theta_1 = \theta_2$, when $\dot{Q} = 0$, which is believed to be true within a few mK when the pressure in the evacuated space is low enough (this assumption was checked experimentally; see below).

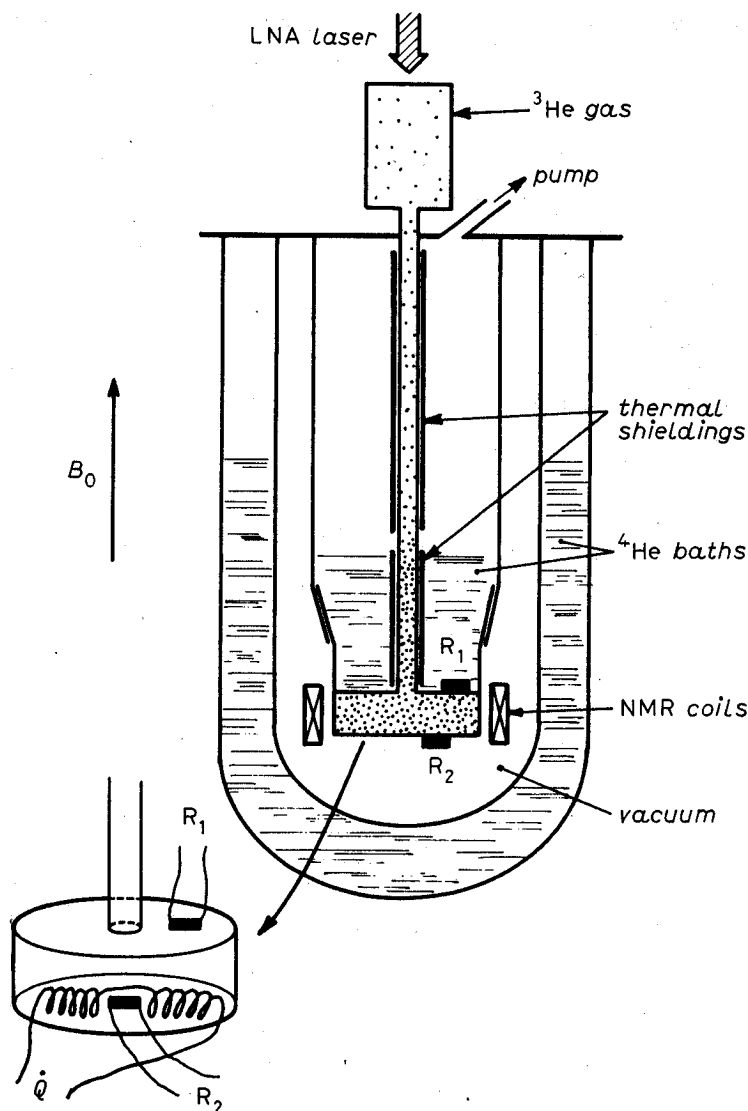


Fig. 1. - Sketch of the apparatus. The ^3He gas under study is contained in a double glass cell, in a homogeneous magnetic field B_0 . A weak gas discharge is maintained in the upper cell at room temperature where the atoms are nuclearly polarized by optical pumping. Spin diffusion towards the lower cell creates the polarization in the lower cell. This polarization is monitored by NMR. The lower cell is also shown at a larger scale, with carbon resistor R_1 and R_2 ; the bottom plate is covered with resistive heating wires.

The magnetization M of the gas in the lower cell is monitored by NMR with a set of crossed coils located in the evacuated space; free induction decay signals at 10 kHz can be detected after small tilting angle pulses in the induction coils. The calibration of the NMR signals (within 10%) is obtained by comparison with optical measurements of the nuclear polarization in the upper cell performed with the method described in [15] (measurement of the circular polarization of the lines emitted by the discharge). This comparison is made in a situation where the nuclear polarization is nearly the same in the two cells.

Some care is necessary for the solid hydrogen coating to play well its anti-relaxing role. During the cooling process, when the hydrogen freezes in the cell, one avoids temperature gradients (which might result in nonuniform thickness of the hydrogen film) by introducing a few Torr of ^4He in the space around the cell (see fig. 1). This gas is, of course, subsequently

evacuated before taking measurements. Under the best experimental conditions, we were able to obtain nuclear polarizations reaching 30%, the limitations being mainly introduced by wall relaxation in the lower cell (local $T_1 \approx 30$ min at 2.5 K but only 3 min at 1.8 K).

As a first step of the experiment, the heat conductivity $\kappa(0)$ of unpolarized ^3He gas was measured. Various values of \dot{Q} ranging from 0 to $100 \mu\text{W}$ were applied and temperatures θ_1 and θ_2 were recorded; this corresponds to temperature differences not exceeding 100 mK, well below the onset of convection in the cell [16]. The difference $\theta_2 - \theta_1$ increases linearly with \dot{Q} and the heat conductivity $\kappa(0)$ is obtained from the slope

$$K(0) = \frac{\dot{Q}}{\theta_1 - \theta_2} \cdot \frac{l}{S}, \quad (2)$$

where l is the spacing between the plates and S their area. Actually, corrections to this formula are needed for two reasons: i) heat conduction through the side walls of the cylindrical glass container and ii) temperature differences between the two faces of each of the plates. More details about these corrections will be given in a subsequent article; correction i) is the dominant one and varies between 40% at 4.2 K and 20% at 1.6 K (where the glass conduction is lower); correction ii) never exceeds 10%. Overall, the values obtained between 1.6 and 4.2 K for the unpolarized gas heat conduction do not differ by more than 5% from earlier results [16, 17].

The main measurements were then performed in the presence of nuclear polarization: starting with a polarized gas at temperature θ_1 , temperature θ_2 is monitored as a function of time with a fixed heat flux (typically $\dot{Q} \approx 100 \mu\text{W}$). Temperature θ_1 is kept as constant as possible (within ≤ 1 mK) during the measurement. Then, the nuclear polarization M is suddenly destroyed by a $\pi/2$ NMR pulse; the subsequent change $\delta\theta_2$ in temperature is observed, as visible in fig. 2; $\delta\theta_2$ is typically 1 or 2 mK when $M \approx 20\%$ or 30% . The corresponding decrease $\delta\kappa$ of the heat conductivity of the gas is finally obtained from formula (2), with corrections included (here, only correction ii) comes in).

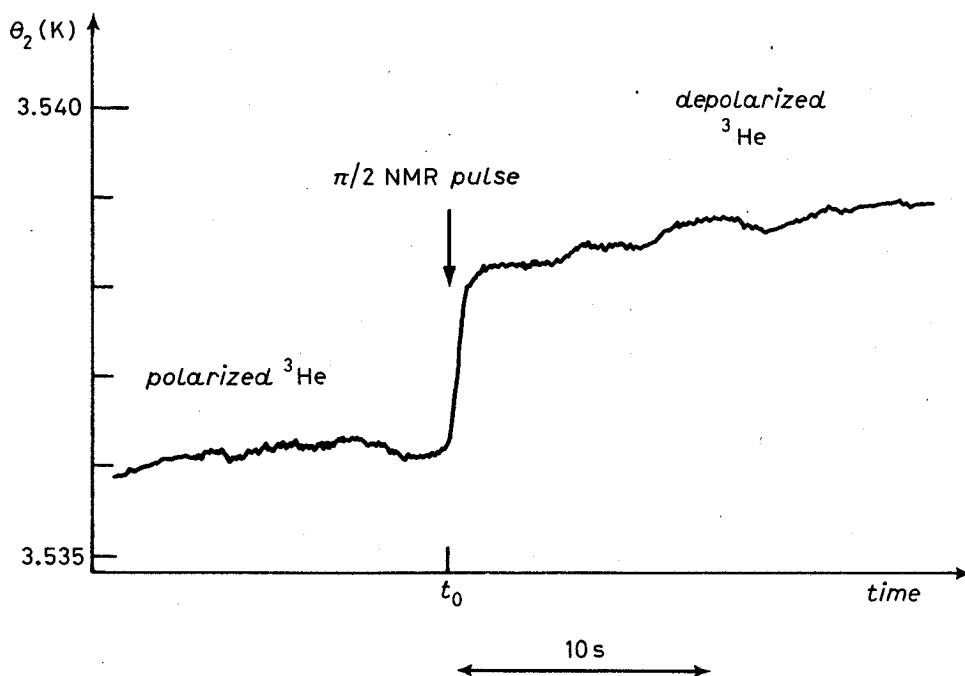


Fig. 2. – Signal delivered by resistor R_2 as a function of time (the vertical scale is given in temperature θ_2). The power \dot{Q} applied to the bottom plate is $107 \mu\text{W}$ and temperature θ_1 of the upper plate is kept approximately constant (≈ 3.430 K). The nuclear polarization, $M = 25\%$, is suddenly destroyed at time t_0 by a NMR pulse. The subsequent raise in temperature θ_2 is due to the quantum effect studied.

We first checked that, at a fixed temperature, $\delta\kappa$ varies quadratically as a function of M , as expected in the range of M available in the experiment ($M \leq 30\%$) [2, 4] (higher-order corrections in M^4 etc. are smaller than 1% under these conditions). Another check was made: if no external heating power \dot{Q} is applied, no temperature change is observed on R_2 when the gas is depolarized by NMR, provided the pressure in the evacuated space is low enough. This shows that no significant spurious heat flux reaches the lower plate and that the calibration of R_2 described above is correct.

The measurements were then carried out at different temperatures. Figure 3 shows the results obtained for the M -independent quantity $\delta\kappa/M^2\kappa(0)$ as a function of $\theta = (\theta_1 + \theta_2)/2$ ⁽²⁾. Each measurement has a statistical error of approximately 5% in relative value; temperature calibration error and spurious \dot{Q} are estimated to introduce smaller errors. If the calibration of M is incorrect, the only correction is a change of the vertical scale.

For comparison, fig. 3 also shows the theoretical predictions of [4], when two different He-He interaction potentials are used. This reference also gives a discussion of the physical origin of the variation of the effect as a function of temperature. Generally speaking, the agreement between theory and experiment is good; nevertheless, the differences, in particular in the positions of the maximum, are sufficiently marked to be significant.

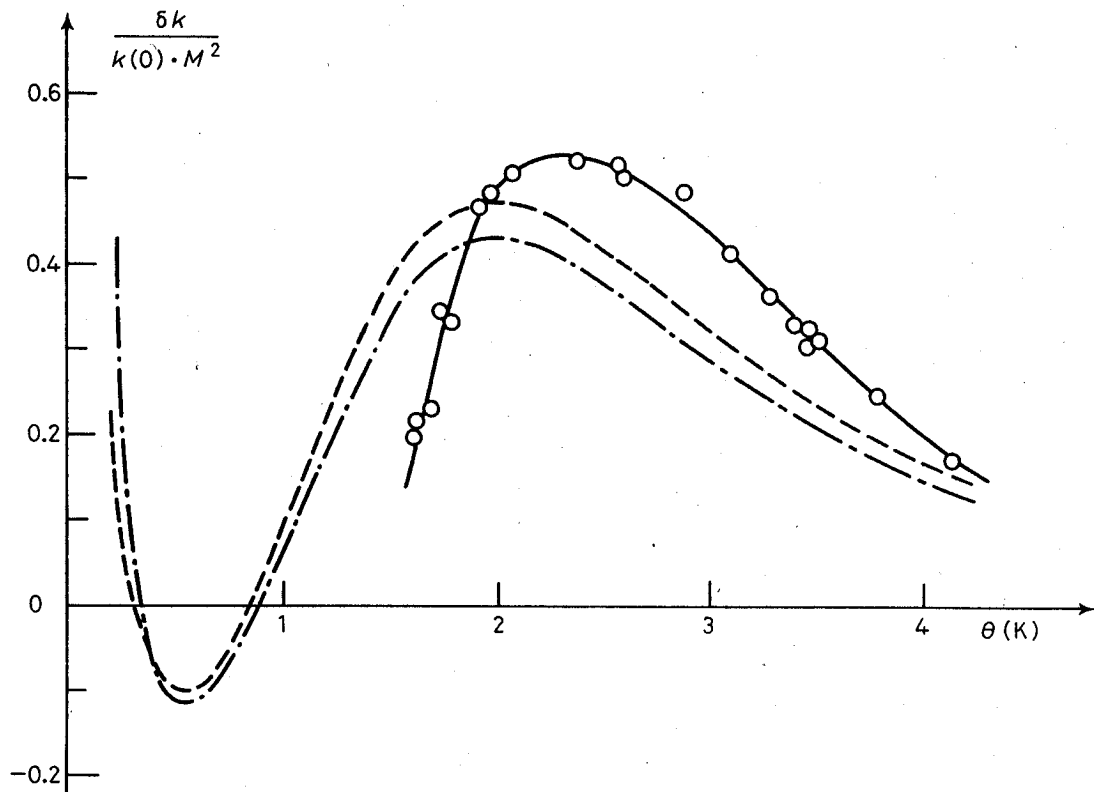


Fig. 3. – Open circles: values of $\delta\kappa/\kappa(0)M^2$ measured as a function of the temperature θ of the gas. The solid line is merely a guide for the eye. For comparison, the predictions of ref. [4] are also shown (dashed line: Lennard-Jones potential; dashed-dotted line: Aziz potential).

In conclusion, our experiments show that the differences between the properties of gaseous $^3\text{He} \uparrow$ and ^3He are clearly visible, with macroscopic detection techniques which do not rely (as NMR does) on the existence of a nonzero nuclear magnetic moment. Even with relatively low nuclear polarizations, never exceeding 30%, the accuracy of the measurements is relatively good; it is superior for example to what was obtained for spin

⁽²⁾ The values of ref. [16, 17], more accurate than ours, were taken for $\kappa(0)$.

waves in ^3He [8]. The theoretical predictions of [2, 4] are confirmed but, interestingly, the experimental results indicate the necessity of improving these calculations. It would be useful to check if higher order Sonine polynomial corrections significantly change the results of [2, 4]. If not, our results show that, as has often been the case in the past, when new physical properties are measured in a gas, new information on the interatomic potential is obtained; in other words, the Aziz helium-helium potential [18] would have to be slightly modified in order to incorporate our data.

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REFERENCES

- [1] KELLER W. E., ^3He and ^4He (Plenum Press, New York, N. Y.) 1969.
- [2] LHUILLIER C. and LALOË F., *J. Phys. (Paris)*, **40** (1979) 239; **43** (1982) 197.
- [3] BASHKIN E. P. and MEYEROVICH A. E., *Sov. Phys. JETP*, **47** (1978) 992; *Adv. Phys.*, **30** (1981) 1.
- [4] LHUILLIER C., *J. Phys. (Paris)*, **44** (1983) 1.
- [5] BASHKIN E. P., *JETP Lett.*, **33** (1981) 8; *Sov. Phys. JETP*, **60** (1984) 1122; MEYEROVICH A., *J. Low Temp. Phys.*, **53** (1983) 487.
- [6] LHUILLIER C. and LALOË F., *J. Phys. (Paris)*, **43** (1982) 225.
- [7] JOHNSON B. R., DENKER J. S., BIGELOW N., LÉVY L. P., FREED J. H. and LEE D. M., *Phys. Rev. Lett.*, **52** (1984) 1508.
- [8] NACHER P. J., TASTEVIN G., LEDUC M., CRAMPTON S. B. and LALOË F., *J. Phys. (Paris) Lett.*, **45** (1984) L-441.
- [9] GULLY W. J. and MULLIN W. J., *Phys. Rev. Lett.*, **52** (1984) 1512.
- [10] GREYWALL D. S. and PAALANEN M., *Phys. Rev. Lett.*, **46** (1981) 1292.
- [11] KOPIETZ P., DUTTA APRIL and ARCHIE C. N., *Phys. Rev. Lett.*, **57** (1986) 1231.
- [12] SCHEARER L. D., LEDUC M., VIVIEN D., LEJUS A. M. and THERY J., *IEEE J. Quantum Electron.*, **22** (1986) 713; DANIELS J. M., SCHEARER L. D., LEDUC M. and NACHER P. J., submitted to *J. Opt. Soc. Am. B*.
- [13] LEDUC M., CRAMPTON S. B., NACHER P. J. and LALOË F., *Nucl. Sci. Appl.*, **2** (1984) 1.
- [14] LEFÈVRE-SEGUIN V., NACHER P. J., BROSSEL J., HARDY W. N. and LALOË F., *J. Phys. (Paris)*, **46** (1985) 179.
- [15] PAVLOVIC M. and LALOË F., *J. Phys. (Paris)*, **31** (1970) 173.
- [16] BETTS D. S. and MARSHALL R., *J. Low Temp. Phys.*, **1** (1969) 595.
- [17] KELLER W. E. and KERRISK J. F., *Phys. Rev.*, **177** (1969) 341.
- [18] AZIZ R. A., NAM V. P. S., CARLEY J. S., TAYLOR W. L. and MCCONVILLE G. T., *J. Chem. Phys.*, **70** (1979) 4330.