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# Spin rotation effects and spin waves in gaseous 3He↑

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# Spin rotation effects
# and spin waves in gaseous ³He↑

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**Résumé.** — Les gaz polarisés comme H↑ ou ³He↑ possèdent des modes oscillants et amortis basse température, les ondes de spin, dont l'origine est l'effet de rotation des spins identiques se produisant lors de collisions entre particules indiscernables. Cette lettre décrit l'observation de ces effets dans ³He↑ dans le domaine de température $2 < T < 6$ K, et des mesures préliminaires du coefficient $\mu$ qui donne la surtension de ces ondes. Ces mesures sont en accord avec les calculs numériques de C. Lhuillier.

**Abstract.** — Spin polarized gases such as H↑ or ³He↑ can propagate damped spin waves at low temperatures; their origin is the so-called « identical spin rotation effect » in collisions between identical particles. We report here the observation of such effects in ³He↑ in the temperature range $2 < T < 6$ K, and preliminary measurements of the coefficient $\mu$ which characterizes the quality factor of the spin oscillations. The measurements are in agreement with the numerical calculations of C. Lhuillier.

Spin polarized helium and (atomic) hydrogen are systems which exhibit a number of unusual and interesting properties at low temperatures [1]. This is true even for dilute polarized gases for which, although degeneracy effects are negligible, the transport properties can be strongly modified by nuclear polarization [2]. Similar phenomena have been predicted by E. Bashkin and A. Meyerovich for solutions of ³He in superfluid ⁴He, in the low temperature regime were they are degenerate [3, 4], and have also been studied by W. Mullin and K. Miyake [5].

Perhaps one of the most striking effects produced by the spin polarization is the existence of spin waves [6, 7]. For a dilute gas of unpolarized atoms the only propagating mode corresponds to sound waves, since heat conduction, viscosity and spin diffusion give rise to purely damped modes. On the other hand, for a polarized gas of identical atoms, the evolution of the spins becomes oscillatory and can be understood in terms of (damped) spin waves. This phenomenon is a pure consequence of particle indistinguishability and arises from the so called « identical spin rotation effect » during collisions [7]. Another difference between sound and spin waves is that the former do not depend crucially on collisions and are obtained in the zero order approximation of hydrodynamics (Euler equations), while the latter appear to first order (Navier-Stokes equations).

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Spin waves are actually well known phenomena in degenerate liquids [8-10]; their existence is a consequence of the anisotropic part of the « molecular field » in the Landau theory of strongly degenerate Fermi liquids. A. J. Leggett has shown how these waves affect the current of spin diffusion in the liquid [11] and has also predicted new effects occurring in spin echo experiments (Leggett-Rice effect [12]). These predictions have been confirmed in experiments with liquid $^3$He at very low temperatures ($T \lesssim 10$ mK) and used to measure the Fermi liquid interaction parameter $\lambda$ for pure $^3$He and $^3$He-$^4$He mixtures [13]. For spin-polarized dilute gases the situation is rather different, and no phenomenological parameter has to be introduced in the theory [7, 14], so that all transport properties (heat conduction, viscosity, spin waves) can be calculated *ab initio* from the interatomic potential ([1]). This has been done recently by C. Lhuillier for H↑. D↑ and $^3$He↑ [15], and the theoretical prediction is that spin waves will strongly modify the properties of spin diffusion of these gases even at relatively high temperatures, of the order of 1 K or more. For this discussion, it is convenient to introduce the (relative) nuclear polarization $M$ of the gas ($-1 < M < 1$), and a dimensionless coefficient $\mu$ which is the ratio between « identical spin rotation cross sections » and the cross section for collisions between distinguishable particles [7]. The product $\mu M$ gives the relative importance of the particle indistinguishability effects in spin diffusion; for example, for a cell where the lowest diffusion mode has a diffusion time $\tau_D$, the frequency of the transverse spin wave is given (in the Larmor rotating frame) by ([2]) :

$$\delta\omega = \frac{\mu M}{\tau_D}. \tag{1}$$

In other words, $\mu M$ is a spin oscillation « quality factor », independent of the gas density $n$ (whereas $\delta\omega$ itself is *inversely* proportional to $n$).

## 1. The experiment.

Measuring the properties of spin waves in spin polarized gases, and in particular their coefficient $\mu$, is therefore interesting for two reasons : observing for the first time a new mode in a dilute gas and, by comparison with numerical calculations [15], obtaining information on the interatomic potentials which is completely independent of the data from other methods, in an energy range where there are relatively few experimental data.

Several experiments of this type are in progress, one with doubly spin polarized hydrogen [17], the other with low concentration solutions of $^3$He in $^4$He which can be seen as non-degenerate gases (but the experiment also includes data in the degenerate regime) [18]. Their results will be published shortly [18, 19].

Here we report on experiments with gaseous $^3$He↑, nuclearly polarized by optical pumping, as well as preliminary measurements of the coefficient $\mu$ as a function of temperature. The waves were excited by applying a gradient of magnetic field over the sample but, unlike the experiments previously done on liquid $^3$He [13], no spin echo technique was used. Instead of creating strong gradients of magnetization over the sample, we have studied a situation where the atoms explore all the volume available inside the container before being significantly affected by the applied magnetic gradient (motional averaging condition [20, 21]); clearly, this is possible only if the walls of the gas container have only a very weak relaxation effect. In this kind of situation, the effects of spin waves can be simply and completely described [22] in terms of the longitudinal

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([1]) For the same reason, these properties can be exactly calculated for any value of the spin polarization $M$, unlike in a liquid where an expansion for small $M$ values is necessary.

([2]) We assume here that $\mu M \ll 1$, so that the blocking of the transverse diffusion, which is second order in $\mu M$, is negligible (in the notation of reference [22], $\tilde{\tau}_D \simeq \tau_D$).

relaxation time $T_1$, the transverse relaxation time $T_2$, and a frequency shift $\Delta\omega$ of the precession of $M_+$ the transverse component of $M$.

The origin of the shift can be understood from the following simple argument, which is valid when $| M_+ |$ is small compared to the longitudinal component $M_0 \simeq M$. If the spin polarization $M$ were strictly uniform over the sample, no shift could ever be introduced by the « identical spin rotation effect » (or the molecular field), simply because of angular momentum conservation [11]. The rôle of the gradient $\delta B_0$ of the longitudinal component of the magnetic field is precisely to create a gradient of spin polarization, that is to couple the total transverse magnetization $M_+$ of the sample to a spin wave. But this wave oscillates with a slightly different frequency ($\delta\omega$ in the Larmor rotating frame), and is also damped at a rate $1/\tau_D$. The situation is then simply that of 2 coupled oscillators with different frequencies and damping, and one can easily show that this coupling introduces for $M_+$ a damping rate $1/T_2$ and a frequency shift $\Delta\omega$ given by :

$$\frac{1}{T_2} \simeq a \mid \delta B_0 \mid^2 \tau_D \tag{2}$$

$$\Delta\omega \simeq a \mid \delta B_0 \mid^2 \tau_D \times \mu M \tag{3}$$

where $a$ is some coefficient which is irrelevant for our discussion (these formulae are valid to second order in the gradient $\delta B_0$ when $\delta B_0 \tau_D \ll 1$ ; we have assumed that the gradient is linear, so that only the lowest diffusion mode is significantly excited). We note that :

$$\Delta\omega \, T_2 = \mu M \tag{4}$$

a relation which we have made use of to measure $\mu$.


## 2. Experimental set-up.

A general sketch of the experimental set-up is shown in figure 1. The $^3$He gas is contained in a sealed pyrex cell, consisting of two different parts connected by a 3 mm i. d. tube : a 5 cm i. d. by 5 cm long cylindrical container where the atoms are submitted to a discharge and optical pumping at ambient room temperature, and a 13 mm i. d. spherical bulb where the N.M.R. experiments are done at low temperatures. Gaseous diffusion inside the connecting tube transfers polarization from the upper cylinder to the lower bulb. With this experimental arrangement optical pumping can be done at a temperature where it is much more effective than at a few degrees Kelvin. The source of light for optical pumping is a colour centre laser operating at $\lambda = 1.08$ μ. The sealed cell contains a mixture of $^3$He and molecular hydrogen, which freezes on the cold parts of the inside walls of the cell. This provides a « cryogenic coating » which strongly reduces the nuclear relaxation [23, 24]. The pressure of the $^3$He gas was $p \simeq 0.3$ torr corresponding to number densities $n \simeq 10^{16}$ and $n \simeq 10^{18}$ cm$^{-3}$ at high and low temperatures, respectively. Thermal shields are used to control the temperature gradient along the tube. Their rôle is to concentrate the thermal gradient to a small region, in order to reduce the area of surfaces at intermediate temperatures (cold enough to relax $^3$He nuclear spins but not cold enough to be covered by a few layers of solid hydrogen). Nuclear polarizations of 30 % or more in the lower bulb can be obtained with a laser power of about 100 mW. More details about laser optical pumping and the polarization transfer technique can be found elsewhere [25, 26].

As shown in figure 1, the measurement bulb is inside a small isolated inner cryostat (i.d. : 5 cm) placed inside the main helium cryostat. In this way, the temperature of the bulb can conveniently be varied between 1.5 and 8 K. In some cases, it was found useful (or even indispensable) to heat this cell to about 8 K for a few minutes, in order to improve the quality of the H$_2$ cryogenic coating and allow polarization transfer. The N.M.R. system has two separate, and orthogonal pairs of induction and detection coils. The latter coils have a diameter of 26 mm, and are wound
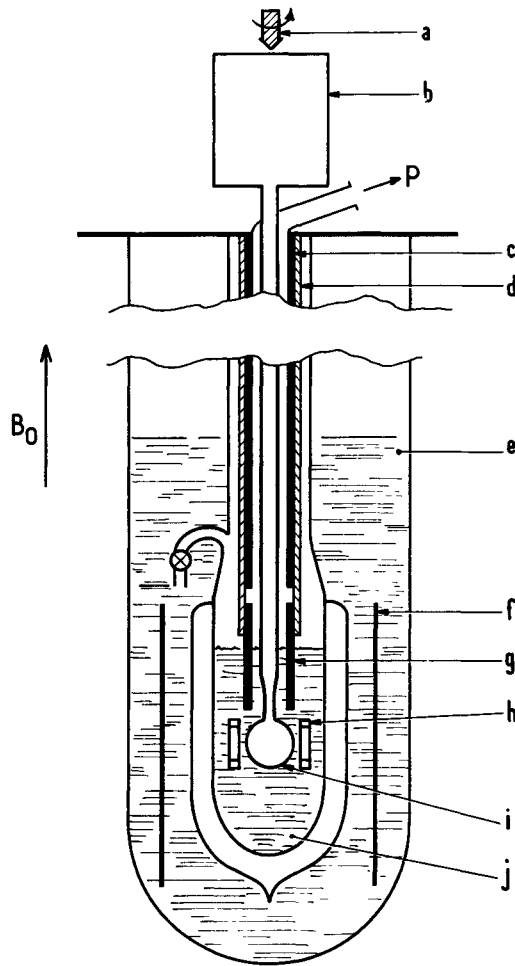
Fig. 1. — Semi realistic sketch of the experiment. a. circularly polarized laser beam ; b. upper ³He container submitted to optical pumping at 300 K ; c. thermal shielding at high temperature ; d. insulating foam ; e. helium bath at 4.2 K ; f. cylinder of superconducting lead ; g. thermal shielding at low temperature ; h. N.M.R. pick-up coils (other sets of coils for tilting the spins and varying the field gradients are not shown) ; i. measurement ³He sample ; j. pumped ⁴He bath at adjustable temperature ; P. to vacuum pump.

with 40 turns each ; we shall discuss later why it was found useful to add in series with the detection coils another inductance not coupled to the spins. The N.M.R. frequency was $\simeq$ 12 kHz, which corresponds to a (vertical) magnetic field of about 4 G. The stability of this field was greatly improved (to about $10^{-7}$ G) by using a vertical superconducting cylinder to trap a fixed flux. This cylinder (length : 22 cm ; diameter : 9.2 cm) was made of brass, and its internal wall was covered by a thin layer ($\simeq$ 1 mm) of lead. The shielding factor obtained in this way was $\simeq 10^4$, in good agreement with theoretical predictions [16]. The field gradient was of the order of 2 mG/cm, depending on how the superconducting cylinder was cooled in the 4 G vertical field produced by 3 horizontal coils (diameter 1 m). By adjusting the currents in 3 sets of compensation coils creating pure gradient of the longitudinal field at the cell, it was possible to reduce the initial gradients by a factor up to 10.

## 3. Measurements.

The quantities measured in our experiments were the precession frequency and the transverse relaxation time $T_2$ of a small transverse component of the nuclear polarization at various positive and negative values of the longitudinal polarization $M$. Reversals of $M$ were accomplished by $\pi$ N.M.R. pulses, and $\pi/20$ pulses were used to sample $M$ in an almost non-destructive way. The amplitudes of responses to $\pi/20$ pulses were converted to polarization measurements by calibrating the detection system with the signal from a small circular coil. Due to the relatively high signal-to-noise, the limit of accuracy of the measurement of $M$ was set by the 5-10 % calibration uncertainty. A consequence of the high magnetization was that, if no special care was taken, a Zeeman maser oscillation [27] started even when the pick-up coils had a low $Q$ factor, $Q \simeq 5$ for example. Even below maser threshold, the radiation reaction of the pick-up coils could easily produce frequency shifts 10 times larger than $\Delta\omega$. The solution of this problem was found in a strong reduction of the filling factor of the circuit. This was obtained by reducing to 40 the number of turns of the pick-up coils and adding in series with them another inductance (a pair of coils with 300 turns each) not coupled to the spins. A simple reduction of the number of turns with no additional inductance would not be effective : a high value for the tuning capacitance $C$ would become necessary to match a low inductance coil, resulting in increased electrical currents in the coils and larger reaction on the spins. Experimentally, it is possible to check that the radiation damping due to the pick-up coils is indeed weak : this is obtained when the measured values of $T_2$ are independent of $M$ (in particular of its sign). The $Q$ factor of the whole circuit was 20 and the amplitude of the spin signal across the capacitor about 1 μV.

With the particular geometry of our cell the time constant for build up of nuclear polarization at 4.2 K in the lower cell was of the order of 20 min. This time was effectively limited by a 1 mm i.d. by 5 cm long capillary tube shrunk to 0.3 mm i.d. over one cm of its length just above the lower cell. Although the capillary increased somewhat the time required to pump up $M$ in the lower cell, it also increased the lower cell confinement time and strongly reduced spurious frequency shifts due to spin diffusion within the connecting tube.

After a sufficient (20-30 %) value of $M$ was obtained, a series of measurements was started. The procedure was to alternate $\pi/10$ pulses to measure $T_2$ and the spin precession frequency, and $\pi$ pulses to reverse $M$. Repeating this operation 5 or 10 times reduced $M$ by a factor $\simeq 5$, so that measurements of the spin frequency for a whole set of positive and negative values of $M$ was obtained (with the additional 300 turn inductance, $T_2$ was found to be practically independent of $M$). The actual measurement was obtained by beating in a lock-in detector the spin free induction decay signal with a stable reference frequency, and recording the phase shift during the decay. The phase shift $\Delta\varphi$ accumulated during a time $T_2$ was found to vary linearly with $M$, as predicted by equation (4); from the slope of this linear variation and a measurement of $M$, the value of the coefficient $\mu$ was extracted.

Typical values were :

$$T_2 \simeq 0.5\text{-}5 \text{ s}$$

$$(\Delta\omega/2\,\pi) \simeq 1\text{-}10 \text{ mHz}$$

which shows that even very small spurious frequency shifts could be a problem in these experiments. The experimental procedure was chosen to eliminate all shifts which do not depend on $M$. There are nevertheless several possible origins for $M$-dependent shifts : N.M.R. coil reaction and spin diffusion in the connecting tube, as discussed above, change of the average field experienced by the gaseous sample due to its own field (non-sphericity of the cell), and magnetic image of the sample in the superconducting shield (this turns out to give a correction of the order of 1 mHz in our experiments). The two last effects are more important at lower temperatures, when the gas density is higher. Fortunately, these frequency corrections do not depend on $\delta B_0$

or $T_2$. To eliminate these effects, we extrapolated the values of $\Delta\varphi/M_0$ to zero $T_2$ values (i.e. infinite $\delta B_0$), as shown in figure 2. The measured points then fall as expected on a straight line having slope compatible with an order of magnitude of estimate of the magnetic image effect. The intersection of the line with the vertical axis gives the value of $\mu$ at the temperature of the sample. Some care was taken to determine experimentally the sign of $\mu$, which requires a know-ledge of the sign of $M$. To this purpose, we removed the series inductance in the detection circuit, and obtained maser oscillation for the orientation having $M > 0$ (if $B_0 > 0$).

The values of $\mu$ obtained in this way are shown in figure 3, as compared to C. Lhuillier's cal-culations (full line) [15]. The agreement within present accuracy is satisfactory and clearly confirms the predicted spin wave behaviour. We think that the precision of the measurements could be improved in the future and the temperature range extended.

## 4. Conclusion.

These experiments show that spin rotation effects can be detected in gaseous ground state $^3$He↑. In our experiments, the spin waves are more strongly damped than in liquid systems [18, 28], or spin polarized hydrogen [17], simply because the product $\mu M$ is larger for those systems. This is why, in our case, only one wave (corresponding to the lowest diffusion mode) has been observed, and not the rich mode structure seen in references [19] and [28]. It is interesting to note that recent experiments [29] on degenerate solutions of $^3$He in superfluid $^4$He between 0.3 and 25 mK seem to indicate that the isolation of a higher mode is also possible. Finally, we remark that, for heat conduction and viscosity, the effects of a spin polarization are more pro-nounced in $^3$He↑ than H↑, because $^3$He is a fermion system. We are therefore planning experi-ments to investigate other transport properties of spin polarized $^3$He. Another interesting exten-sion would be to perform experiments below 2 K, where an appreciable fraction of a $^3$He mono-layer can be formed, and study transport properties in two dimensional systems [30].
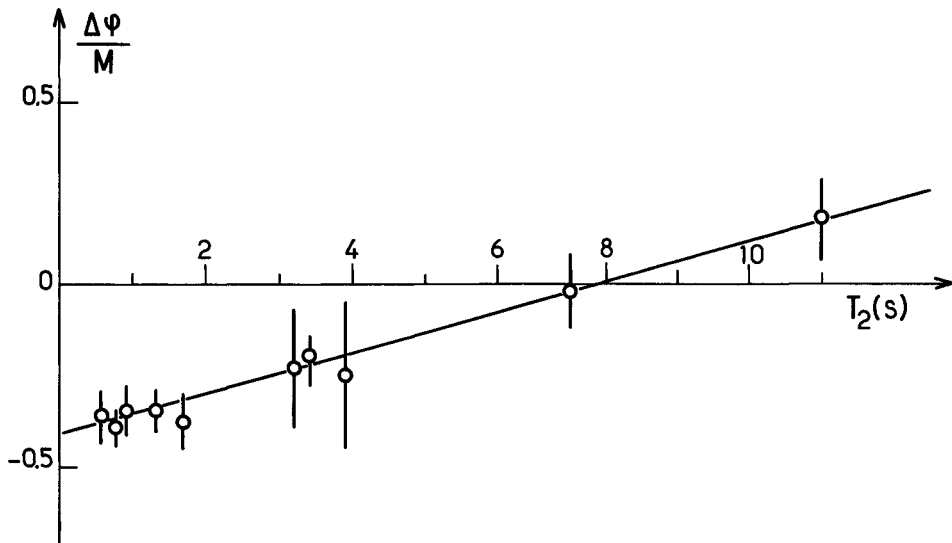


Fig. 2. — Measurements at 2.5 K. The horizontal scale is the relaxation time $T_2$ of the transverse nuclear polarization. $1/T_2$ is proportional to the square of the field gradients over the sample. The vertical axis gives the phase shift $\Delta\varphi$ (measured during a time $T_2$) divided by the spin magnetization $M$.
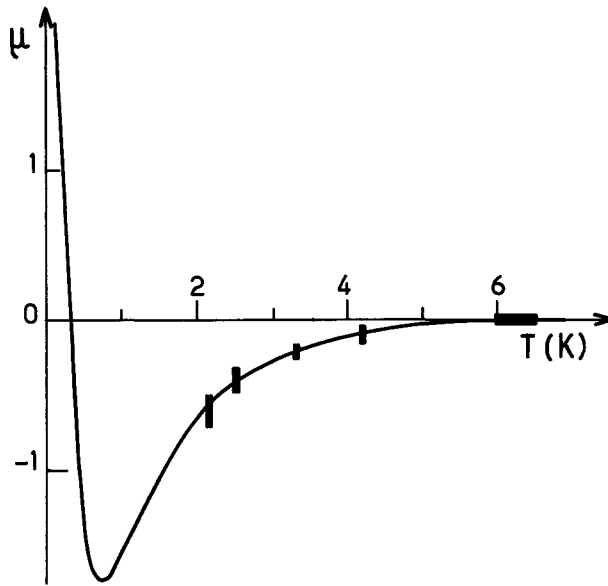
Fig. 3. — Values of the coefficient $\mu$, characteristic of spin rotation effects in ³He↑. The solid curve shows the theoretical predictions of [15]. The points are the results of this experiment.

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