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Very Long ^{3}He Nuclear Relaxation Times at 4 K Using Cryogenic Coatings

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Longitudinal ^{3}He nuclear relaxation times at low temperature have been measured by an optical-pumping technique, using solid H_2 coatings on the cell walls. Relaxation times longer than 2 days at 4.2 K were obtained with a 3-cm-diam cell. Preliminary results with other coatings (solid D_2, Ne, etc.) are reported.

Although the first optical-pumping experiment was performed with an atomic beam,[1,2] it is often better to confine the atoms, ions, or molecules under study in a closed cell. This leads to more intense and narrower resonance lines. The problem then is to find a suitable transparent container so as to minimize the effect of atom-wall collisions on the relaxation times of the relevant atomic levels. In the case of alkalis for example, the use of internally-paraffin-coated cells can strongly reduce the effectiveness of disorienting wall collisions, leading to relaxation times of the order of 1 sec[3,4] and very narrow magnetic-resonance curves.[5] In a similar way, Teflon coatings have given good results for the hyperfine relaxation in the ground state of hydrogen.[6,7] With a given coating, going to higher temperatures usually lengthens the relaxation time. This is because the atom dwell time on the walls gets shorter, which makes the disorienting interaction less efficient. In this way, nuclear relaxation times as long as 10^3 sec have been obtained with ^{199}Hg and ^{201}Hg.[8]

Another example which has been extensively studied is the nucleus of ^{3}He ($I=\frac{1}{2}$). It can be strongly polarized (up to about 20%) by optical pumping of the helium 2^3S_1 metastable states produced in a weak discharge, and metastability exchange transfer of the orientation to the ground state.[9] At room temperature, and in the absence of the discharge, the ground-state longitudinal relaxation time T_r of ^{3}He atoms is typically 2 h.[9] As it is mainly due to permeation of the atoms through the Pyrex,[10] one can use special nonpermeable glasses and get T_r values as long as 2 days[11-13] (or even longer at higher temperature[11]). At 77 K, T_r remains very large, and the nuclear orientation is still high.[11,14] But, when the temperature decreases, since the dwell time of the atoms on the walls becomes longer and longer, T_r becomes very short,[15] preventing a sizable nuclear orientation buildup (moreover, orientation transfer via metastability exchange becomes less efficient[16]). As a consequence, in optical-pumping experiments at very low temperature (4 K for example), one easily observes the magnetic resonance of the 2^3S_1 metastable state,[17] but not the ground-state resonance. However, in many respects, it would be interesting to operate at such low temperatures. This would allow the reduction of the effect of collisions in the gas; it might facilitate the production of a higher density of oriented ^{3}He nuclei for a polarized target.

We report here some results concerning the use of wall coatings which allow very long relaxation times and sizable nuclear orientation of ^{3}He at 4 K. Instead of using paraffin or Teflon coatings (which could be destroyed by the discharge), we filled the sealed cell, not only with ^{3}He, but also with another nondiamagnetic gas which, when cooled, solidifies on the walls and provides the desired coating (hereafter called cryogenic coating). Most experiments were done with Pyrex spherical cells 3 cm in diameter (a few with 6 cm diameter) sealed at room temperature, containing a few torrs of ^{3}He gas and, in some cases, additional gas coatings (Ne, Ar, Kr, Xe, H_2, D_2, and CH_4). A classical silvered-glass cryostat was used to maintain the cell in liquid helium. Detection of the longitudinal nuclear polarization in the ground state was obtained by measuring the circular polarization of the 6678- and 5876-Å helium lines emitted by the discharge; the principle of this sensitive detection method (rotating $\lambda/4$ plate allowing lock-in amplification) has been described elsewhere.[18]

We measured first T_r for uncoated cells, by monitoring the pumping buildup time of the nuclear polarization. With a 6-cm Pyrex cell, we found a very abrupt variation: At 30 K, $T_r \simeq 15$ sec (instead of a few hours at 77 K) and at 21 K, T_r goes down to the minimum measurable value $T_r \simeq 1$ sec. At 4 K, T_r is probably equal to the diffusion time of ^{3}He atoms to the walls ($T_r \simeq 10^{-2}$ sec) and no nuclear-orientation signal was observed, as found in Ref. 11.

With cryogenic coatings, T_r can be so large that another measurement sequence was used. After formation of the coating by cooling the cell to 4.2 K, the sample is polarized by the discharge optical-pumping technique.[9] When the stationary nuclear polarization is obtained, the discharge and the light beam are turned off; ^{3}He nuclear spins then freely relax during a time interval Δt which varies from a few seconds (practically equivalent to $\Delta t = 0$ since T_r is very long) to a few days. The discharge is then turned on again and one immediately measures the remaining nuclear polarization $P(\Delta t)$. A semilogarithmic plot of $P(\Delta t)$ versus Δt gives a straight line, whose slope provides the value of T_r. Experimentally, we found a good linear fit, and T_r was measured with an accuracy better than 15%. No systematic variations of T_r, with the same cell, but with coatings formed during successive cooling cycles, were observed.

In our experiments, the effect of the magnetic relaxation was not negligible; this relaxation is due to the diffusion of ^{3}He atoms through the weak magnetic field gradients over the cell volume produced by the laboratory environment (the gradient due to the coils was negligible since the relative inhomogeneity of their field was $\lesssim 10^{-5}$). The corresponding longitudinal relaxation time T_m is proportional to ω_0^2 [9,19,20] when $\omega_0\tau_d \gg 1$ [21] (ω_0 is the static applied magnetic field in angular-fre-

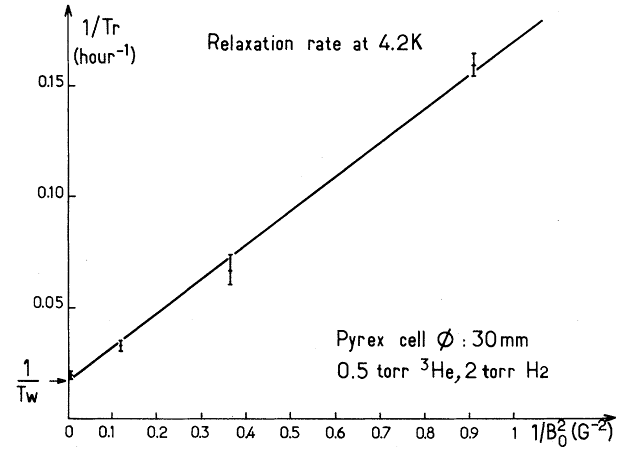

FIG. 1. Variation of the nuclear relaxation probability for an H_2-coated cell, as a function of the inverse of the square of the magnetic field. The extrapolated value at infinite field gives the wall relaxation time $T_w \simeq 60$ h.

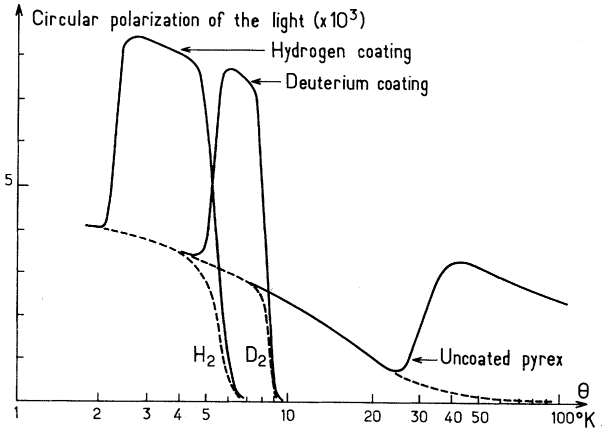

FIG. 2. Polarization of the optical lines emitted by the He discharge as a function of the temperature for coated and uncoated cells. Full lines: NMR off; dashed lines: NMR on (no ground-state polarization). For the bare Pyrex cell, no ground-state polarization is observed under 21 K. With an H_2 coating, one observes a sizable nuclear polarization between 2.5 and 6 K. At higher temperature, the solid H_2 coating disappears and the H_2 pressure in the cell becomes too large to permit any ^{3}He orientation by optical pumping. With a D_2 coating, the same phenomena are observed at higher temperatures.

quency units and τ_d is the diffusion time of ^{3}He atoms in the cell). The total relaxation probability $T_r^{-1} = T_m^{-1} + T_w^{-1}$ is thus a linear function of ω_0^{-2}. Figure 1 shows the measured values of T_r^{-1} in various magnetic fields for a spherical cell (diameter $2R = 3$ cm), filled at room temperature with 0.5 Torr of ^{3}He and 2 Torr of H_2 (coating of about 30 molecular layers of solid H_2). One gets a straight line whose extrapolation when $\omega_0^{-2} \rightarrow 0$ gives an intrinsic relaxation time $T_w = 60 \pm 10$ h. T_w can be ascribed to the effect of atom-wall collisions, since the bulk relaxation time T_b due to dipole-dipole interaction is very long for our low-density samples.[9,22,23] The solid-hydrogen coating produces then a tremendous improvement of the wall's qualities ($T_w \simeq 10^8 \tau_d$). Since the average wall collision frequency $3\bar{v}/4R$ for a given atom is about 10^4 sec^{-1} ($\bar{v}$ is the mean atomic velocity), 2×10^9 wall collisions are required for flipping a nuclear spin.

At the present time, we have not yet made a detailed study concerning the T_w dependence on the coating thickness. A cell filled with 0.17 Torr of H_2 (mean coating thickness of 3 molecular layers) gave a relaxation time too short to be measured ($T_r < 1$ sec). This result may be due to an incomplete coating of the walls. It is also remarkable that, except H_2, none of the other mentioned coatings (in particular D_2) led to a measurable value of T_w at 4.2 K.

We have only preliminary results concerning the relaxation times on these coatings at temperatures higher than 4 K. We simply measured, during the temperature rise of the cryostat, the optical-pumping efficiency, i.e., the circular polarization of the lines emitted by the discharge. Such experiments do not provide precise values of T_w, and the measured signal is in fact sensitive to T_w only when 1 sec $\lesssim T_w \lesssim$ 100 sec (if $T_w < 1$ sec, no nuclear orientation signal is observed; if $T_w > 100$ sec, the wall relaxation is masked by the other discharge relaxation processes). Furthermore, the measured circular polarization does not depend on the ground-state orientation alone, but also depends on the metastable orientation, because of direct electronic excitation of atoms from the 2^3S_1 state.[24] In order to distinguish between the contributions of the ground state and 2^3S_1 levels, the optical circular polarization was measured in the presence and in the absence of a saturating magnetic resonance in the ground state. Figure 2 shows the results obtained between 1.8 and 40 K with an uncoated cell, and with H_2 and D_2 coatings.

Without coating, no ground-state orientation is observed below 21 K, and the measured polarization only comes from the metastable orientation. This polarization decreases from 4 to 21 K, under the effect of the more and more effective metastable relaxation due to metastability exchange collisions with unpolarized ground-state atoms.[16] Above 21 K, T_r approaches the minimum measurable value of 1 sec, and the polarization grows, as a result of the contribution of the ground-state orientation. With the H_2-coated cell, one clearly sees that this coating gives good relaxation times T_r between 2 and 6.5 K (above this temperature, the H_2 coating disappears and the H_2 pressure in the cell becomes too high for optical pumping to work). Similarly, a D_2 coating is good in the range between 6 and 8.5 K. We also tried CH_4 coatings and obtained a measurable ground-state orientation between 5 and 40 K (but the measurements can be perturbed by the breaking up of CH_4 due to the discharge, leading to molecular H_2). Finally, we used Ne, Ar, Kr, and Xe coatings; preliminary results indicate that they allow sizable nuclear orientations in the ranges 8–13, 14–36, 16–50, and 17–33 K, respectively.

It seems that the differences between the relaxation properties of H_2 and D_2 coatings are related to the 4-K interval between their melting curves. However, more precise measurements are obviously needed for a complete interpretation of the various properties of these coatings, and further experiments using a temperature-controlled cryostat are planned. Several applications of the present work can be considered, for example, orientation of liquid ^{3}He by optical pumping.[25] The achievement of relaxation times longer than a week at 4 K (with 6-cm-radius cells) might be useful in the search for an upper limit of the ^{3}He electric dipole moment.[26,27] Finally, it appears that cryogenic coatings are suitable for optical-pumping studies of the properties of clean and well-defined surfaces.

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