Pulse NMR of $^3$He in aerogel at temperature 1.5 K

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Abstract. Experimental data of $^3$He pulse NMR in contact with aerogel (95%) in a nuclear Larmor frequency range of 5 to 20 MHz at $T=1.5$ K are reported. For the first time spin kinetics of $^3$He (adsorbed, gaseous and liquid) has been studied systematically at temperatures higher than degeneracy temperature of this quantum liquid. It has been shown, that in all cases the nuclear spin-lattice relaxation time $T_1$ is linearly proportional to the Larmor frequency, while the spin-spin relaxation time $T_2$ is frequency independent. The magnetic relaxation times $T_1$ and $T_2$ are linearly proportional to the ratio of the magnetization of whole spin system to magnetization of the adsorbed layer of $^3$He.

1. Introduction

Aerogel consists of a network of randomly interconnected solid particles of amorphous SiO$_2$, 3-5 nm in size with an average distance between strands of about 50 nm. Because of open geometry and large porosity (in a present work a sample of aerogel 95% porosity has been used), as well as a large surface area (as much as 1000 m$^2$/g), aerogel is a unique system for the study of impurity and surface effects.

Superfluid $^3$He in aerogel is studied extensively, and by a wide variety of methods, but little information exists on the nuclear magnetic relaxation of either the degenerate and nondegenerate Fermi liquid $^3$He (The degeneracy temperature is about 0.5 K ) in aerogel [1,2]. The spin kinetics of $^3$He in different phases in confined geometry is also a matter of interest [3,4].

The goal of present work was to determine impact of different mechanisms on the nuclear magnetic relaxation of $^3$He in contact with aerogel. The experiments were all done at a temperature of 1.5 K.

2. Experimental part

The spin kinetics of $^3$He in contact with aerogel has been studied using pulse NMR techniques in a Larmor frequency range between 5 MHz and 20 MHz.

A 95% porous aerogel sample (d=5 mm, h=7 mm) was sealed leak tight in a glass tube. Gaseous $^3$He was condensed in the sample at temperature 1.5 K. A home-made pulse NMR spectrometer was used to measure the nuclear magnetic relaxation. Spin-lattice relaxation times were measured using the "saturation-recovery" method, by measuring the free induction decay (FID) amplitude after the saturating pulse and $\pi/2$-pulse. Spin-spin relaxation times were measured by Hann method. The temperature 1.5 K was achieved by pumping on a $^4$He bath.

The saturation of aerogel by $^3$He was carried out in the following sequence: gaseous $^3$He was condensed in the experimental cell in small amounts (about 0.5 cm$^3$ STP). The pressure in the system...
was monitored, and when it reached a value less than $10^{-2}$ torr the next dose of $^3$He was added. When the equilibrium pressure in the system rose above $10^{-1}$ torr, the entire surface of the aerogel was assumed to be covered by $^3$He. Further condensation of $^3$He led to an increase of the pressure with most of the $^3$He remaining in the gas phase. When the pressure approached the saturation value (50.3 torr at 1.5 K), the liquid phase appeared and further dosing of $^3$He was controlled by the $^3$He NMR signal amplitude.

3. Results
In all experiments, with $^3$He adsorbed on the surface, adsorbed and gaseous $^3$He as well as with adsorbed and liquid $^3$He present in the aerogel, $T_1$ and $T_2$ showed behavior that could be characterized by a single exponential. No separate NMR signals that could be attributed to mobile (liquid and vapor) and a localized (i.e. adsorbed) $^3$He were observed. This can be explained by fast molecular exchange in the system.

![Figure 1](image1.png)

**Figure 1.** Frequency dependence of spin-lattice relaxation time $T_1$ and spin-spin relaxation time $T_2$ in aerogel at $T=1.5$ K: ○ – adsorbed $^3$He, ▲ – adsorbed and gaseous $^3$He (25 torr), ■ – adsorbed and liquid $^3$He

![Figure 2](image2.png)

**Figure 2.** The dependence of spin-lattice relaxation time $T_1$ and spin-spin relaxation time $T_2$ on the amount of $^3$He in the aerogel, $f_0=10$ MHz, $T=1.5$ K.

In all cases the spin-lattice relaxation time $T_1$ linearly depends on the $^3$He Larmor frequency (Fig.1a), while the spin-spin relaxation times in all phases of $^3$He in aerogel is independent of the
NMR frequency (Fig.1b). In Fig.2a and Fig.2b the dependence of $T_1$ and $T_2$ on the amount of $^3$He in the system is presented. As can be seen, these dependences are linear.

To further investigate the influence of the adsorbed $^3$He film on the magnetic relaxation of $^3$He, experiments have been conducted in which the aerogel surface was coated with (non-magnetic) $^4$He. The amount of $^4$He necessary to cover the sample surface was determined by the same methods as for $^3$He. After the aerogel surface was covered by a layer of $^4$He, $^3$He was added until the equilibrium pressure reached 25 torr. This coverage of the aerogel surface slows down the magnetic relaxation by more than an order of magnitude (Fig.3a and Fig.3b).

![Figure 3.](image)

Figure 3. The $^3$He longitudinal magnetization recovery and transverse magnetization decay curves of gaseous $^3$He in aerogel with the surface covered by: ○ $^3$He, ● $^4$He, both at an equilibrium $^3$He pressure of 25 torr and $T$=1.5 K.

4. Discussion

The linear frequency dependence of $T_1$ and the fact that $T_2$ is independent of frequency in adsorbed $^3$He layer (Fig.1a and Fig.1b) may be explained on the basis of Cowan’s mechanism for magnetic relaxation for $^3$He in 2-D films [5]. This mechanism is dependent on quantum exchange in the solid adsorbed layer of $^3$He. Cowan introduced the conception of 2-D spin-diffusion with a characteristic frequency $\omega_c$. At some condition on a correlation function in frames of this theory calculations give linear dependence of $T_1$ on frequency in frequency range $0.3<\omega/\omega_c<3$. For explanation of linear dependence of $T_1$ on frequency for substrate with non-homogenous surface, for instance DLX-6000 powder, by Maegawa et al [6] has been proposed the idea about existence of $\omega_c$ frequency spectra, which corresponds to localization of $^3$He in cavities with different surface area and linear frequency dependence of $T_1$ exists in a wide frequency range. It is not clear to what extend this theory, which was constructed for purely 2-D systems, is applicable to the fractal structure of aerogel.

The measurements of $T_1$ and $T_2$ versus amount of $^3$He in the system (Fig.2a and Fig.2b) show, that both $T_1$ and $T_2$ are linearly proportional to the relaxation times in the adsorbed layer and the magnetization of the entire spin system scaled by that of the atoms in the adsorbed layer, i.e.:

\[
T_1 = T_{1S} \cdot N_0 / N_S, \tag{1}
\]

\[
T_2 = T_{2S} \cdot N_0 / N_S, \tag{2}
\]

where $T_1$ and $T_{1S}$ are the spin-lattice relaxation times, $T_2$ and $T_{2S}$ the spin-spin relaxation times, and $N_0$ and $N_S$ are the number of spins for the entire spin system and for adsorbed layer respectively.

Surprisingly, the transition from a system consisting of a gas phase plus an adsorbed layer, and one that contains liquid has no effect on the magnetic relaxation rates of the spin system as a whole. The amount of $^3$He in gaseous and liquid states was estimated by comparison of FID amplitude in these
phases and the FID amplitude of the adsorbed layer. From Fig.2a and Fig.2b can be seen, that amount of $^{3}$He in the adsorbed layer is about 9% of maximum amount of $^{3}$He in aerogel.

These results can be explained by the magnetic reservoir model of Hammel and Richardson [7]. According to this model the entire $^{3}$He magnetization relaxed via the adsorbed, ("localized"), $^{3}$He on the substrate surface with the process of magnetization transfer between "localized" and "mobile" spins taking place faster than $T_{1S}$.

Experiments with the aerogel surface coated with nonmagnetic $^{4}$He support the idea that the magnetic relaxation of "mobile" (liquid and gaseous) $^{3}$He takes place via the immobile surface layer of $^{3}$He. Magnetic relaxation times rises more than on order of magnitude (Fig.3a and Fig.3b) at the same $^{3}$He pressure.

The non-exponential behavior of the $^{3}$He transverse magnetization decay in the system with a $^{4}$He coating (Fig.3b) is due to a non-homogenous magnetic field $H_0$ and the existence of an additional mechanism for transverse magnetization decay.

5. Conclusions
Summarizing all experimental data, we can conclude, that the nuclear magnetic relaxation of the entire $^{3}$He spin system occurs through the $^{3}$He layer adsorbed on the aerogel. The intrinsic relaxation mechanisms in the liquid and gas phases is much weaker than that in the adsorbed layer. To explain the observed nuclear magnetic relaxation in the adsorbed layer a new theoretical model is needed that is applicable for the aerogel structure.

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