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Probing the dynamics of ^{3}He atoms adsorbed on MCM-41 with pulsed NMR

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Abstract. We report measurements of the nuclear spin-spin and spin-lattice relaxation times for ^{3}He adsorbed on MCM-41 for temperatures $0.08 < T < 1.2$ K. Deviations from Curie behavior are observed at low temperatures. The relaxation times exhibit a two-component behavior representing the differing dynamics of the mobile quasi-free molecules in the center of the tubes compared to the adsorbed layer on the walls. The amplitudes of the two components provide an accurate measure of the number of fluid-like molecules traveling in the center of the nanotubes.

1. Introduction

Interesting new quantum states have been predicted for quantum fluids (^{3}He, ^{4}He, H_2, HD) constrained to nanoscale dimensions where the de Broglie wavelength and/or the Fermi length become comparable to or larger than the available pore or channel size[1–6]. 1D ^{4}He superfluidity has been reported for low density ^{4}He in nanotubes[4, 7] and the onset of degeneracy has been seen for ^{3}He in the hexagonal channels of FSM16[8, 9]. The mesoporous structure of MCM-41 is similar to that of FSM-16 but with small changes in dimensions. The typical pore size for MCM-41 is 2-4 nm in diameter and 300 nm in length[5, 10]. Previous research[11] has observed the crossover from 2D motion to 1D motion for adsorbed ^{3}He atoms. However, to observe the predicted 1D Luttinger liquid behavior, much lower temperatures are necessary[12]. Studies of ^{3}He in these mesoporous structures to temperatures below 50mK provide a framework that allows the exploration of the unique properties of quantum liquids or solids in confined states.

We have used pulsed NMR techniques to measure the dynamics of 1.08 monolayers of ^{3}He on MCM-41 for temperatures $0.08 < T < 1.2$ K. The nuclear spin relaxations rates are very sensitive to the motion of the ^{3}He atoms and it is possible to distinguish the different components of the NMR signal according to slow and fast motions.

2. Experimental considerations

The MCM-41 sample was loaded into a polycarbonate NMR cell with modest pressure ($\sim$ 10 N/m^2 against a silver end cap that formed the end of an extension from a nuclear demagnetization refrigerator. The NMR probe consisted of a single coil tuned and matched to 50Ω at the cell site and connected to a room temperature hybrid tee bridge[13, 14]. Fine tuning was carried out using small variable capacitances at room temperature to correct for small changes when the NMR probe is cooled. The sample was pumped to a high vacuum and flushed with ^{4}He gas several times to purge adsorbed gases. Fig. 1 shows a typical ^{3}He isotherm

measured at 2.5 K. The volume of the NMR cell is small and with the long ($\sim$ 2 m.) input capillary the accuracy of the isotherm is limited. Nevertheless, the compressibility deduced from the derivative given in the inset to Fig. 1 does show the minima attributed in Ref. [11] to the wall monolayer coverage and pore completion. The first minimum corresponds to 9.5 $\pm$ 1.5 atoms per $(\mathrm{nm})^2$ comparable to the values reported by Taniguchi *et al.*[8].

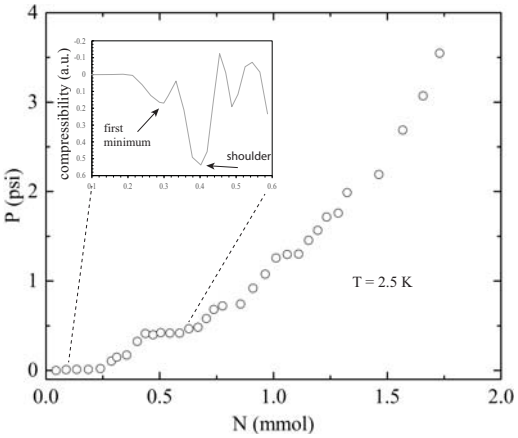

Figure 1. Isotherm of ^{3}He on MCM-41 in NMR cell at 2.5 K. The inset shows the compressibility derived from the isotherm with the characteristic two minima near wall completion.

For the NMR studies gas was admitted to cover the wall plus an additional 9.0% that would be expected to be free to travel inside the nanotubes. The NMR measurements provide a very reliable measure of the fraction of the ^{3}He atoms that are mobile as the nuclear spin-spin relaxation times T_2 of the atoms bound to the wall are very different to those for mobile atoms. The motion of the atoms modulates the nuclear spin-spin interactions and the spin-spin relaxation times are proportional to the frequency of the atomic motion, leading to a long T_2 for the mobile atoms compared to those bound to the wall. This difference is shown in Fig. 2 from which we deduce that the mobile ^{3}He atoms form 0.77 $\pm$ 0.05 % of the total sample, much smaller than expected from the amount of ^{3}He added. A similar observation was made by Taniguchi *et al.*[15] in studies of ^{3}He on FSM-16. They attributed the difference to the formation of an amorphous solid ^{3}He layer on top of the the wall coating similar to the behavior observed by Golub and Pobell[16] for ^{3}He in vycor, leaving a very small gas component in the center.

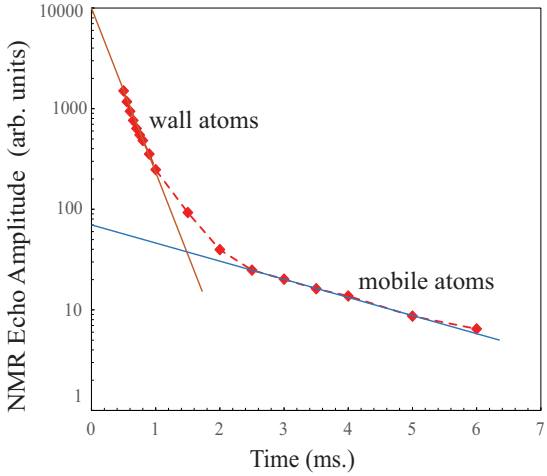

Figure 2. Time dependence of NMR echo decays for ^{3}He in MCM-41 showing the difference in relaxation for the mobile ^{3}He atoms (slow decay) compared to the relatively immobile (fast decay) atoms bound to the walls of the MCM-41.

The nuclear spin magnetization M was measured as a function of temperature using $(\pi/2 - \tau - \pi)$ echos with $\tau = 250\mu s$. A small deviation from Curie's law was observed but

is believed to follow the observation of Taniguchi *et al.*[15] who showed that the amorphous layer can have a broad distribution of very large exchange interactions (up to $J \sim 200$mK) for which the antiferromagnetic susceptibility has pronounced peaks for $T \sim J$[17].

3. Results

The nuclear spin-spin relaxation times, T_2, were measured by following the echo decay of $\pi/2 - \tau - \pi$ RF pulse sequences with variable τ and long waiting times between sequences. A distinctive two-component relaxation was observed for almost all temperatures (see inset of Fig. 4), and the echo decay could not be fit with an ordinary power law decay. The behavior is attributed to a slow relaxation expected for the quasi-fixed atoms adsorbed at the wall and the long-time relaxation component ($\sim 0.8\%$) to atoms free to travel axially along the MCM-41 channels. The mobile fraction is much smaller than expected from the amount of ^{3}He adsorbed. As discussed above this is attributed to the observation of Taniguchi *et al.* that a layer of amorphous solid ^{3}He is formed on top of the layer of wall atoms. The solid black line represents the best fit to an expected $T^{3/2}$ temperature dependence for the long time relaxation for $T > T_F$, where T_F is the Fermi temperature. At low temperatures, $T < T_F$, a linear dependence is expected and this leads to a small departure from the $T^{3/2}$ dependence observed for $0.2 < T < 0.8$K. One does not expect any significant temperature dependence for the short-time relaxation but there is small increase with temperature.

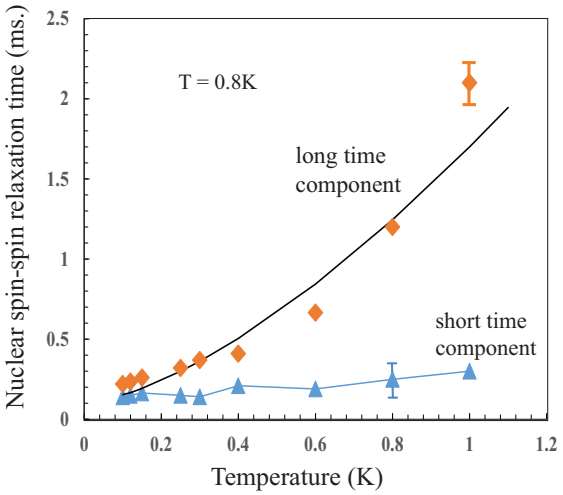

Figure 3. Temperature dependence of the nuclear spin-spin relaxation times for ^{3}He on MCM-41; orange diamonds long-time component, blue triangles short-time component. Typical error bars are shown.

The nuclear spin lattice relaxation rates were determined from repetitive $\pi/2-\tau-\pi$ RF pulse sequences and observing the recoveries to equilibrium. Figure 5 shows the observed temperature dependence for the same sample used for the T_2 measurements. The solid black line corresponds to the expected $T^{3/4}$ dependence deduced from the T_2 behavior with no adjustable parameters.

4. Discussion

In this experiment the nuclear spin relaxation is determined by modulation of the nuclear dipole-dipole interactions between the mobile atoms and the atoms coating the wall, rather than by collisions in the nanochannel. The correlation time τ_c can be estimated from the diffusion constant D using $\tau_c^{-1} = D/a_0^2$ where a_0 is the lattice spacing of the wall atoms. The temperature dependence is given by that of $D(T) = \frac{1}{2}\langle V^2\rangle\tau_{coll}$ where τ_{coll} is the time during the hard core collisions of the mobile atoms. We therefore need to evaluate the mean energy $\langle E\rangle = \int Eg_{1D}(E)F(1-F)dE$ where $g_{1D}(E) = 4n/(EE_F)^{1/2}$ is the 1D density of states and F

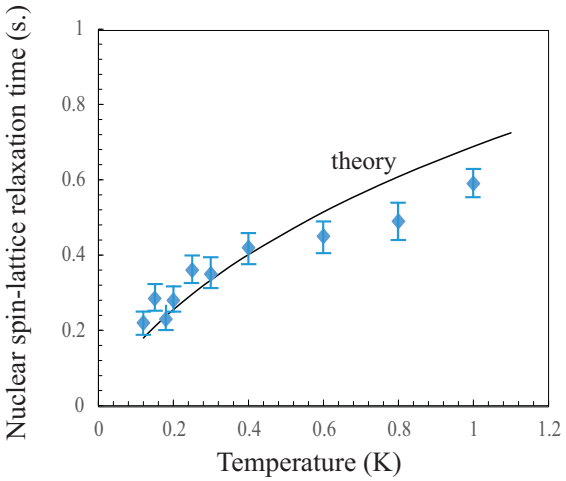

Figure 4. Temperature dependence of the nuclear spin-lattice relaxation time for ^{3}He on MCM-41. A typical error bar is shown. The solid line is the dependence calculated from the T_2 temperature dependence of Fig. 3 without any adjustable parameters.

is the Fermi function. We find $\tau_c^{-1} = Ana_\perp \frac{k_b}{\hbar} \frac{T^{3/2}}{T_F^{1/2}}$ for $T > T_F$. n is the linear density, $a_\perp$ is the transverse dimension and A is a numerical constant. The nuclear spin-spin relaxation time is given by $T_2 = M_2^{-1}\tau_c^{-1}$ where M_2 is the NMR second moment for the ^{3}He-^{3}He wall interactions. We find $T_2 = BT^{3/2}$ where B is a constant $\sim 10^{-3}$ in excellent agreement with Fig. 4.

As noted by Yager *et al.*[11] the correlation function $g(t) = (t/\tau_c)^{1/2}$ in the hydrodynamic limit and the nuclear spin lattice relaxation time T_1 is therefore given by $T_1^{-1} = 10.5 M_2 (\frac{\tau_c}{\omega_L})^{1/2}$. This relation predicts a $T^{3/4}$ temperature dependence in agreement with the observations shown in Fig. 5. Furthermore, we have $T_2/T_1 = 10.5/(\omega_L\tau_c)^{1/2}$ independent of M_2.The solid line in Fig. 5 was determined from the best fit of Fig. 4, using $\tau_c = 8.3\,10^{-2}T^{-3/2}$ s. The agreement with the predictions for a 1D Fermi gas is excellent. For higher dimensions the temperature dependencies for T_2 and T_1 would have opposite trends, with one increasing while the other decreases with temperature.

It should be noted that the relaxation we observe here is very different from the spin drag relaxation for Fermi gases[18–20] for which $\tau_D \propto T$ at very low temperatures ($T < T_F$) and $\propto T^{-(1/2)}$ at high temperatures ($T > T_F$) with a peak near $T = T_F$.

5. Conclusion

NMR studies of ^{3}He constrained to the interior of nanochannels of MCM-41 have shown the onset of degeneracy below 220 mK. The nuclear spin-spin relaxation has two components corresponding to atoms in the wall and atoms in the center of the tube. The temperature dependence above 220 mK is consistent with that expected for 1D dynamics in the classical regime. Future experiments are planned using ^{4}He and HD plating and measurements at lower temperatures to test for the dynamics predicted for a 1D degenerate Fermi system.[12, 21–23]

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