Journal of Low Temperature Physics (2019) 196:308–313
https://doi.org/10.1007/s10909-018-02123-0

Nuclear Spin Relaxometry of ^{3}He Atoms Confined in Mesoporous MCM-41

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Received: 9 July 2018 / Accepted: 9 December 2018 / Published online: 2 January 2019

Abstract
MCM-41 is a unique mesoporous material with cylindrical-like pores that provide an ideal platform to study 1D properties of adsorbed atoms (Boninsegni and Moroni in J Low Temp Phys 118(1):1, 2000). In this research, we use pulsed NMR to measure the nuclear spin-lattice relaxation times (T_1) of ^{3}He adsorbed onto ^{4}He-preplated MCM-41 over a wide range of temperatures for a line density that would lead to a Fermi temperature of 200 mK. The temperature dependence of the nuclear spin-lattice relaxation time for $0.025 < T < 3.5$ K exhibited a pronounced peak at 195 mK, qualitatively similar to that calculated for the spin relaxation of 1D Fermi systems.

Keywords Luttinger liquid · Helium three · NMR

1 Introduction

There is great interest in studying the fundamental properties of 1D systems where strong correlations between neighbors make all excitations collective and must be described in terms of a Tomonaga–Luttinger liquid (TLL) state [1–4]. Experimental tests for TLL physics have become a major activity in modern condensed matter physics [5]. ^{3}He adsorbed onto mesoporous materials that contain nanoscale channels is of special interest as it can provide an ideal model system for studying 1D physics of uncharged systems. The ^{3}He systems are intrinsically very pure, the density can be varied continuously, and NMR methods can be used to explore the dynamics over a wide temperature range.

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Wada et al. [6–8] have observed 1D ^{4}He superfluidity in nanotubes [9], and the onset of degeneracy has been reported for pure ^{3}He in the hexagonal channels of FSM-16 [10,11]. Yager et al. [12] have confirmed the characteristic $t^{1/2}$ time dependence of the spatial correlation functions studied in low-frequency NMR experiments down to $1K$. Here we report the results of NMR studies at higher frequencies and at the lower temperatures needed to measure the microscopic dynamics [13] characteristic of the Luttinger regime. Astrakharchik and Boronat [14] have calculated the phase diagram for low-density 1D Fermi systems in terms of the Luttinger parameter K and the line density n. K is given by the ratio of the Fermi velocity to the sound velocity. They identified three distinct regimes: (1) a Fermi gas with dominant attractive interactions for $1.0 < K < 1.5$; (2) a super-Tonks–Girardeau regime [15] (repulsive interactions) for $0.5 < K < 1.0$; and (3) a quasi-crystalline regime for $0.0 < K < 0.5$ at high line densities.

2 Experimental Methods

We used commercial hexagonal MCM-41 [16], a material that consists of a remarkably regular array of unidimensional, hexagonal mesopores [17]. The samples of MCM were heated under vacuum to drive off adsorbed water, air and other gases before assembly in the NMR cell. The amount of MCM-41 placed in the cell was 0.2 g, corresponding to an expected surface area of $200 \pm 20\,\mathrm{m}^2$. The real surface area was determined in situ (i.e., while loaded in the NMR cell) from the adsorption isotherms [18]. This monolayer coverage deduced from the fist step in the isotherm was found to be 0.38 ± 0.02 mmol, which is in good agreement with the mass of the MCM-41.

In earlier studies [18] ^{3}He was added to completely coat the walls of the MCM-41 with a small additional amount of ^{3}He added to provide a dilute 1D gas component. The NMR results of those studies confirmed the presence of a diffusive Fermi gas component at high temperatures. In this study ^{4}He gas was added to form a monolayer on the MCM, and then ^{3}He was added, corresponding to 10% of the amount of ^{4}He in the wall layer. From the known cross-sectional area of the hexagonal tubes of MCM-41 and the amount of ^{3}He added, the estimated line density is about 0.10 $(\text{Å})^{-1}$.

The MCM-41 was contained in a polycarbonate NMR cell and lightly packed against a solid silver post that was an integral part of a link to the end of the refrigerator (Fig. 1). The NMR coil was matched to a 50 Ω cable using two capacitors located near the NMR cell. The cable was connected to one port of a hybrid tee that formed an RF bridge (Fig. 2) [19,20] for NMR detection at 50 MHz. The sample coil is impedance-matched to 50 Ω to balance the conjugate ports of the hybrid tee and during the RF pulses provide a null at the RF output.

Standard RF pulse sequences were used to measure T_1 and T_2: $\pi/2 - \tau - \pi$ to produce an echo at time 2τ. Varying the pulse sequence with varying repetition rates was employed to measure T_1, and sequences with variable τ were used to measure T_2. Low-power long-width pulses were used to reduce the heating of the sintered silver, and this limited the accuracy of T_2 measurements.

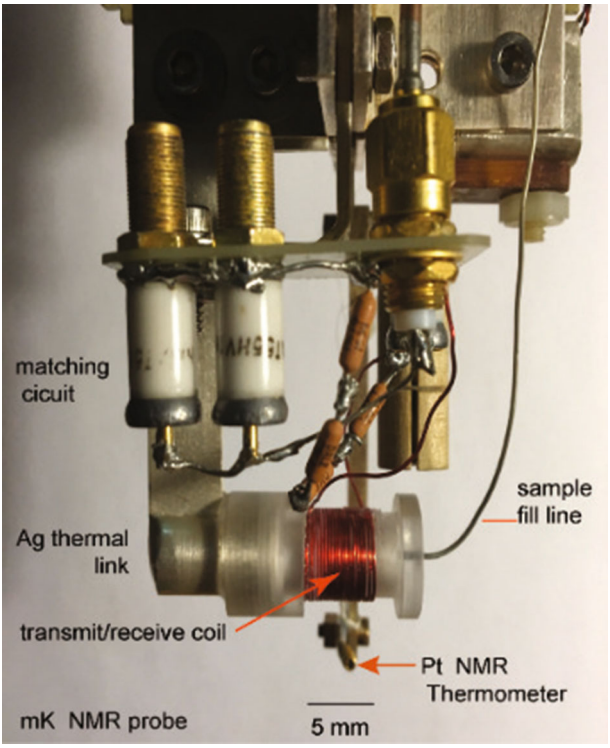

Fig. 1 Picture of NMR cell showing the in situ tuning and matching capacitors (Color figure online)

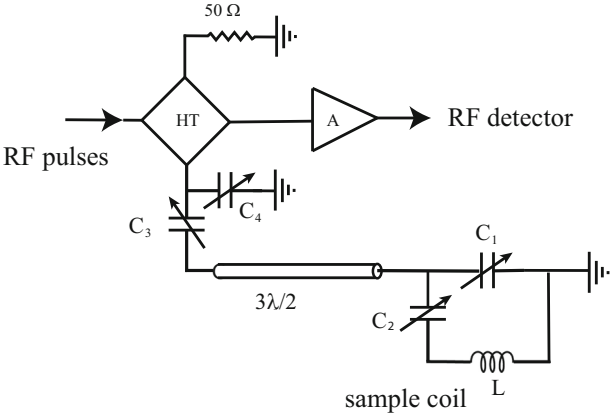

Fig. 2 Schematic representation of RF circuit for the NMR probe, showing use of a 50 Ω stripline hybrid tee (HT) as an RF bridge. C_1 and C_2 match the sample coil L to 50 Ω, and C_3, C_4 correct for any change in the matching when the probe is cooled to low temperatures

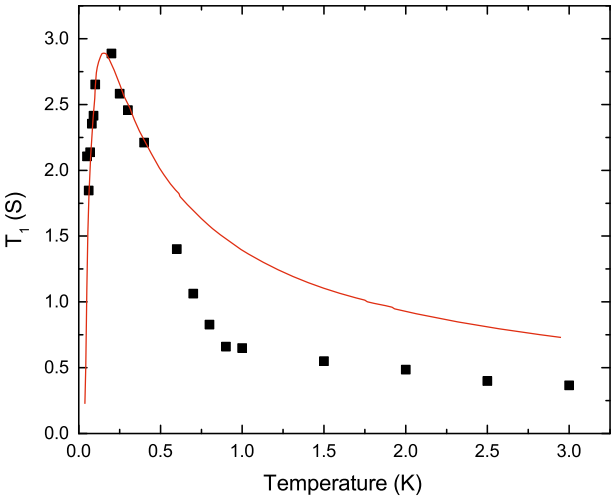

Fig. 3 Temperature dependence of nuclear spin-lattice relaxation time of ^{3}He in MCM-41 for a line density of 0.10 Å^{-1}. The solid red line is the variation expected using the theory of Polini et al. [21,23] (Color figure online)

3 Results

The temperature dependence of the nuclear spin-lattice relaxation times observed for the dilute sample of ^{3}He confined in plated MCM-41 is shown in Fig. 3. A sharp peak is observed at 195 mK and is identified as $2T_F$ [21]. The deduced $T_F = 98$ mK is a factor of two lower than calculated, and this is attributed to a line density of about 30% less than expected from the ^{3}He added. This deficiency is similar to the loss of ^{3}He to sites external to the mesopores identified by Bossy et al. [22] in their neutron scattering studies. Since T_F is proportional to n^2, this can lead to a reduction in T_F by 50%. A linear dependence $T_1 \propto T$ is observed for $T < 2T_F$, and a strong decrease with temperature for $T > 2T_F$.

These observations are to be compared with the results of Polini et al. [21] who predicted the temperature dependences for the spin relaxation rate given by

$$\tau_s^{-1} = \frac{8}{9\pi}\gamma^2 \frac{T}{2T_F}\frac{E_F}{\hbar} \quad \text{for} \quad T < 2T_F \tag{1}$$

and

$$\tau_s^{-1} = 16\pi^{-7/2}\gamma^2 \left[\frac{T}{2T_F}\right]^{-1/2} \frac{E_F}{\hbar} \quad \text{for} \quad T > 2T_F \tag{2}$$

where γ is the dimensionless Yang parameter [23] $\gamma = 2a_{3D}/a_\perp^2 n \sim 1$. a_{3D} is the 3D scattering length, $a_\perp$ is the transverse free dimension of the plated nanotube, and n is the line density. Polini et al. [21] used numerical calculations to determine the full behavior near $2T_F$. They called the relaxation a "spin-drag" response determined from calculations of the dynamic susceptibility.

Note that in high magnetic fields there is a non-negligible nuclear spin polarization and this has been shown to modify the linear low-temperature behavior [23]. For the current experiments the highest polarization reached is 8% at the lowest temperatures and this effect will not be observable.

As shown by the solid line in Fig. 3, while there is good agreement for the low-temperature behavior, the observed relaxation decays faster (almost as T^{-1}) at high temperatures. For very high temperatures we expect the relaxation time to approach a temperature-independent value determined by ^{3}He–^{4}He exchange. This behavior would not explain the deviation from the theoretical $T^{-1/2}$ decay. It should also be noted that the temperature dependence is very different from that observed for bulk sold ^{3}He in the same temperature range where exponential temperature dependencies can be observed with T_1 values in the range 100–1000 s [24].

4 Conclusion

The temperature dependence of the nuclear spin-lattice relaxation times for ^{3}He confined to the interior of ^{4}He-plated MCM-41 shows a distinctive peak at 195 mK, corresponding to a Fermi temperature of 98 mK. This deduced value of T_F is a factor of two lower than that calculated for the line density expected if all the ^{3}He is located in the nanotubes. The discrepancy is attributed to a significant fraction of the ^{3}He being adsorbed in areas outside the centers of the mesopores. Further experiments need to be carried out for different densities and down to lower temperatures at higher magnetic fields where new effects are expected at higher nuclear spin polarizations.

Acknowledgements This research was carried out at the National High Magnetic Field Laboratory's High B/T Facility which is supported by NSF Grant DMR 1644779 and by the State of Florida. This project was also supported in part by an award from the Collaborative Users Grant Program of the NHMFL. We also acknowledge useful discussions with Dmitrii Maslov.

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