



Temperature-linear spin-spin relaxation rates of one-dimensional ^3He fluid formed in nanochannelsTaku Matsushita ^{1,*}, Ryosuke Shibatsuji,¹ Katsuya Kurebayashi,¹ Kazunori Amaike,¹ Mitsunori Hieda ², and Nobuo Wada¹¹*Department of Physics, Nagoya University, Chikusa-ku, Nagoya 464-8602, Japan*²*College of Liberal Arts and Sciences, Tokyo Medical and Dental University, Ichikawa 272-0827, Japan*

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We have made systematic nuclear magnetic resonance measurements for ^3He fluid formed in ^4He -coated nanochannels including the quantum-mechanically one-dimensional (1D) region, which is a possible candidate for a different type of Tomonaga-Luttinger (TL) liquid. Corresponding to the previous heat capacity measurements, a dimensional crossover into the 1D state at low temperatures was observed by the susceptibility. In the 1D region, increases of the spin-spin relaxation time inversely proportional to temperature were found, which is apparently similar to T -linear electron spin resonance linewidths for 1D spin chains. While the relevance for TL liquids is still controversial since the behavior is observed in both degenerate and nondegenerate fluids, increases of the spin-spin relaxation time were confirmed to be observed only when 1D conditions for ^3He fluid are satisfied, indicating that it is a characteristic property of the 1D ^3He fluid.

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In three dimensions or two dimensions, the physical properties of a fluid consisting of interacting fermions are well described by the Landau Fermi liquid theory in which single-particle-like quasiparticles play a main role. In one dimension, however, the situation is expected to change drastically. The strong correlation between neighbor particles necessarily makes all excitations collective; thus, one-dimensional (1D) quantum systems are described as the Tomonaga-Luttinger (TL) liquid instead [1]. While theoretical studies of 1D systems have a long history from individual works by Tomonaga and Luttinger [2–4], experimental studies were later because of the difficulty in making 1D systems with real materials. While the power-law correlation characteristic of a TL liquid appeared to be reported in 1D organic conductors from the 1980s [5], evidence of the spin-charge separation was shown much more recently in quantum wires [6,7]. Although the TL properties represented by the power law have been reported in various 1D systems such as carbon nanotubes [8], edge states in quantum Hall systems [9], quantum spin chains [10], and cold atoms in an optical lattice [11], the comprehensive experimental test of 1D TL physics is still an ongoing issue in condensed-matter physics.

For helium quantum fluids, realization of 1D systems has been attempted using confinement in nanometer-sized channels. Recently, 1D superfluid responses observed in ^4He adsorbed in nanochannels [12–14] have attracted interest for the possibility of being interpreted as those of a bosonic TL liquid [15,16]. For a fermionic 1D fluid, dilute ^3He atoms floating on a ^4He film precoated in nanochannels are a candidate to realize the TL liquid. For such ^3He in nanochannels, a dimensional crossover into the quantum-mechanically 1D state has been shown by the specific heat maximum followed by a decrease of the specific heat at low temperatures [17,18]. The 1D ^3He fluid has several properties different

from electron systems. It has uniform densities but no lattice or interchain exchange. A fluid with no charge enables us to study the effects of interactions other than the Coulomb potential. In addition, the interaction between atoms is suggested to be controlled by the channel size and coated ^4He film thickness [19]. It is theoretically predicted that the combination of repulsive hard-core and attractive van der Waals potentials between ^3He atoms can show various 1D fluid phases with the reentrant density dependence of the Luttinger parameter [20]. Thus, 1D ^3He could be a model system for a TL liquid. Recently, 1D classical diffusion of dilute ^3He in ^4He -coated nanochannels was confirmed by the nuclear magnetic resonance (NMR) above 1.5 K [21]. For relatively dense ^3He , low-temperature NMR was also performed [22,23]. In this Letter, we show the results of systematic NMR measurements performed for dilute ^3He in the 1D state, down to the degenerate regime. The 1D condition was determined from the dimensional crossover shown in the susceptibility. Properties characteristic of a 1D ^3He fluid were investigated by the spin relaxations.

In this study, nanochannels of folded sheet mesoporous materials (FSMs) [24] are employed as a template to form 1D ^3He . FSMs are a family of nanoporous silicate powder with a honeycomb array of straight nanometer-sized channels. The length of the channel is the same as the grain size, about 300 nm, and the channel diameter is uniform and selectable in synthesis. The properties of ^4He adsorbed in the channels were confirmed to change systematically depending on the diameters, which were 1.5 nm (C8) to 2.8 nm (C16) and 4.7 nm [25,26]. Thus, FSM(C14) with a diameter of 2.4 nm was chosen for this study. The FSM powder was packed in a sample cell made of Stycast 1266 epoxy, so that channel axes were randomly oriented against the applied magnetic field. The effective surface area in the cell was measured by the Brunauer-Emmett-Teller (BET) method using the nitrogen adsorption isotherm at 77 K, and areal densities of adatoms were estimated using this surface area. In the superfluid

*matsushita@cc.nagoya-u.ac.jp

experiment [14], the surface area out of these channels was estimated to be less than 3%, so that NMR signals of ^3He other than in channels can be neglected. Film conditions consisting of ^4He and ^3He in these channels were clarified in preceding studies. The channel wall was initially covered with a ^4He film before adsorption of ^3He atoms. Film growth of ^4He in FSM channels was investigated by vapor pressure and heat capacity measurements [25,27]. In general, the effective film thickness estimated from the vapor pressure linearly increases with coverage at first, indicating uniform formation of ^4He film on the channel wall. And above a coverage n_f , adatoms start to fill the void central region in channels. In the film region below n_f , the two-dimensional (2D) compressibility shows a broad minimum at a density n_1 , showing the first atomic layer completion. For the 2.4 nm FSM(C14) used here, n_1 were estimated to be 16 and 18 $\mu\text{mol}/\text{m}^2$ for ^3He and ^4He atoms, respectively, from the heat capacities [27]. The upper limit n_f of ^4He film growth was indicated to be $1.7n_1$ for this channel. The lowest coverage n_c for the 1D superfluid response was observed to be about $1.33n_1$ using a torsional oscillator [14]. Therefore, $1.4n_1$ ($25.0 \mu\text{mol}/\text{m}^2$) of ^4He film between n_c and n_f , where the ^4He film is partially fluid, was prepared for this study, so that the gaslike mobility of ^3He atoms adsorbed on the ^4He surface is guaranteed.

The experimental condition to realize 1D ^3He in ^4He -coated nanochannels was examined with a model of noninteracting gas for the interpretation of the previous heat-capacity study using FSM(C16) and FSM(C12) [18]. In this model, single-particle energy states of ^3He atoms are described as $\epsilon = \hbar^2 k^2 / 2m_3 + \Delta_{lm}$, where m_3 is a mass of ^3He atoms, k is a continuous wave number for atomic motion along the channel axis, and Δ_{lm} is a discrete energy with quantum numbers l and m for excitations of radial and azimuthal motions, respectively. The lowest excitation is Δ_{01} for azimuthal motion, which is estimated to be subkelvin as $A\hbar^2 / 2m_3 (2\pi/\pi d')^2$, where $A = 1-8.9$, depending on the strength of the adsorption potential and d' is the channel diameter narrowed by the precoated ^4He film. When the Fermi energy of ^3He atoms is larger than Δ_{01} , excited states with azimuthal motion are partially occupied even at $T = 0$. Therefore, ^3He must be dilute to realize a quantum-mechanically genuine 1D fluid, which is typically a few percent of n_1 . In this study, ^3He with densities of $0.04-0.95 \mu\text{mol}/\text{m}^2$, corresponding to $0.25\%-6\%$ of n_1 or line densities of $0.15-3.6 \text{ nm}^{-1}$, were adsorbed in the ^4He -coated channels. In addition, the temperature T has to be lower than Δ_{01}/k_B to neglect thermal excitation of azimuthal motions. In the 1D state, even though the ^3He wave function expands in the whole channel cross section, the hard core of about 0.3 nm in diameter is still smaller than the channel radius. Therefore, exchange between ^3He atoms is considered to occur via tunneling through a potential barrier necessary for shrinkage of the wave packets.

For this system, pulsed NMR for ^3He atoms was performed at 4.29 MHz down to 60 mK. The NMR single coil was thermally anchored to the middle of the heat exchanger in the dilution refrigerator to avoid warming up the sample by Joule heating. The signal was measured by a phase-sensitive detector and several ten signals were averaged to improve the signal-to-noise ratio. The susceptibility χ was measured by extrapolation of the free-induction decay (FID) to the

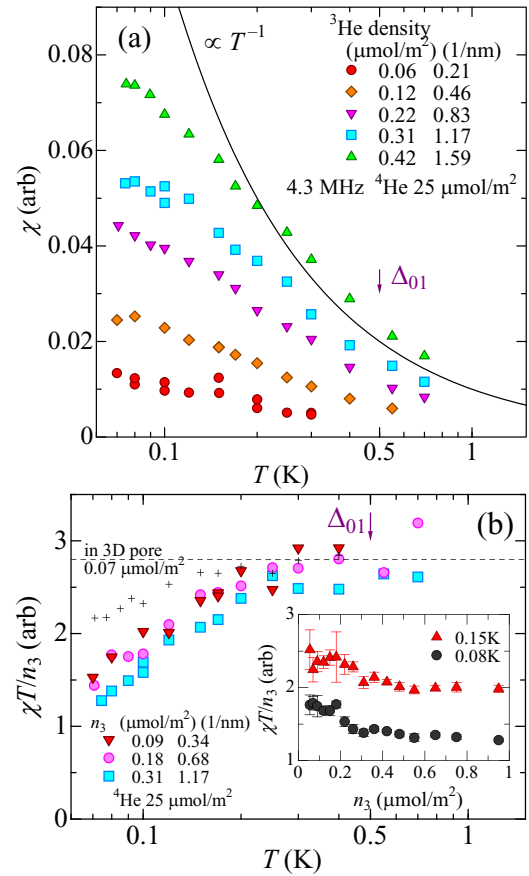


FIG. 1. (a) Temperature dependences of the nuclear magnetic susceptibility χ of ^3He in 1D channels with various ^3He densities n_3 , derived by fitting the form $I(t) = I_0 \exp(-t/T_2^*)$ to the free induction decay after a $\pi/2$ pulse. (b) Temperature dependences of $\chi T/n_3$ for relatively low n_3 . The dashed line is a guide for the eyes corresponding to Curie's law. For reference, crosses indicate $\chi T/n_3$ of ^3He adsorbed in 3D interconnected nanopores coated with a $25 \mu\text{mol}/\text{m}^2$ ^4He film. The inset shows isotherms of $\chi T/n_3$ for ^3He in 1D channels.

time of the rf pulse. The longitudinal relaxation time T_1 was measured by the ordinary comb-pulse method using a pulse sequence with comb- t_1 - 90° - t_2 - 180° - t_2 -echo. For the spin-spin relaxation time T_2 , the spin echo after 90° and 180° pulses was measured. In this system, application of a field gradient did not show further spin echo decay due to spin diffusion, so a field gradient of about 10 G/cm was applied in the T_2 measurement to suppress FID. The absence of diffusion effects also suggests that the possible ^3He diffusion range in the T_2 timescale is not longer than a few times the channel length at low temperatures [28].

Figure 1 shows susceptibilities χ of ^3He at various ^3He densities n_3 . Preliminary low- n_3 data were shown in Ref. [29]. In the specific heat, a dimensional crossover into the 1D state is indicated by a decrease in the gas constant R of a 2D Boltzmann gas and the accompanying Schottky-like peak, whose temperature is almost n_3 independent at $\sim \Delta_{01}/2k_B$ [17,18]. In contrast to the specific heat, χ of the Boltzmann gas is known to obey the same Curie law independent of the dimensionality. However, χ in one di-

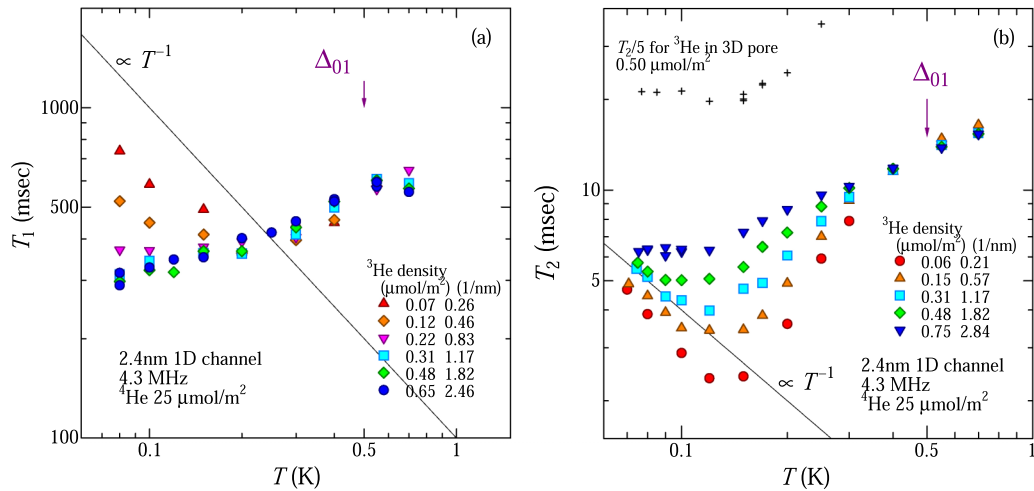


FIG. 2. Temperature dependences of nuclear spin relaxation times for various ${}^3\text{He}$ densities n_3 in 1D channels. (a) Longitudinal relaxation times T_1 . The function $I(t) = I_\infty - I_0 \exp(-t/T_1)$ was fitted to the relaxation curve, where I_∞ is the intensity observed without comb pulses. For fitting, only data with intensity $I \leq I_\infty - 0.5I_0$ were used to ignore a longer thermal relaxation. (b) Spin-spin relaxation times T_2 . The function $I(t) = I_0 \exp(-t/T_2)$ was fitted to spin echoes with the intensity $I \geq 0.15I_0$. For reference, T_2 of ${}^3\text{He}$ ($T_F \approx 0.18$ K) adsorbed in 3D connected nanopores coated with ${}^4\text{He}$ of $25 \mu\text{mol}/\text{m}^2$ are shown by crosses.

mension becomes significantly smaller by a degenerate effect even at temperatures much higher than the Fermi temperature T_F . As a result, the dimensional crossover for nondegenerate ${}^3\text{He}$ can be detected by a reduction from the Curie law, around $T \sim \Delta_{01}/2k_B$, independently of n_3 , which was shown in Ref. [29] with a simulation result using the dimensional crossover model explaining the heat capacity. Figure 1(b) shows χT normalized by n_3 , which is constant for the Curie susceptibility. As a reference, χT of ${}^3\text{He}$ in three-dimensional (3D) nanopores of HMM-2, whose pore size and absorption potential are similar to those of FSM channels [30], is also shown. For dilute ${}^3\text{He}$ in 1D channels, reductions from the Curie law similarly occur below about 0.25 K higher than that in 3D pores, indicating the dimensional crossover. Isotherms in the inset show that they are almost identical for n_3 below $0.2 \mu\text{mol}/\text{m}^2$. Thus, the lowest excitation energy Δ_{01} for azimuthal motion in this channel is found to be around 0.5 K. As shown in Fig. 1(a), T -independent susceptibilities characteristic of degenerate states were observed only in a narrow n_3 region between 0.31 and $0.36 \mu\text{mol}/\text{m}^2$ below 0.1 K because of the overlapping dimensional crossover effect. Recalling that the constant χ appears below $T \sim 0.5T_F$ in a 1D gas, the Fermi temperature $T_F = (\hbar^2/2m^*k_B)(\pi N/2L)^2$ is considered to be about 0.2 K for $n_3 = 0.31 \mu\text{mol}/\text{m}^2$. In this case, the effective mass m^* is estimated to be $1.4m_3$, which is reasonable for a hydrodynamic mass of ${}^3\text{He}$ on a ${}^4\text{He}$ film, compared with the other ${}^3\text{He}$ system floating on the ${}^4\text{He}$ surface [31]. In addition, the value $\Delta_{01}/k_B \sim 0.5$ K is also appropriate for $m^*/m_3 = 1.4$, which was discussed previously in Ref. [29]. Assuming N^2 -linear dependence of T_F for a 1D gas, T_F should exceed Δ_{01}/k_B at $n_3 = n_{\text{lim}} \sim 0.5 \mu\text{mol}/\text{m}^2$. Above n_{lim} , azimuthal excited states are occupied even at $T = 0$, so that the system does not have a quantum-mechanically 1D region. As shown later, the corresponding qualitative change is observed in the spin-spin relaxation above n_{lim} , which supports the validity of this T_F estimation. Thus, “genuine” 1D

conditions for ${}^3\text{He}$ in this channel have been determined as $T < \Delta_{01}/k_B - T_F$ and $n < n_{\text{lim}}$.

Nuclear spin relaxation times for ${}^3\text{He}$ in this system are shown in Fig. 2. For longitudinal relaxation at low T , especially below 0.15 K, a tail continuing over 10 s became remarkable in addition to the exponential form [28]. Since this is attributed to thermal relaxation of the sample after an rf pulse, the relaxation time of the initial part is shown in Fig. 2(a) as a longitudinal relaxation time T_1 . At $T > 0.4$ K, where azimuthal motion of ${}^3\text{He}$ is thermally excited, both T_1 and spin-spin relaxation times T_2 shown in Fig. 2(b) decrease monotonically with decreasing T . They can be regarded as spin relaxations via dipolar coupling at $\omega\tau < 1$, where ω is the Larmor frequency and τ is the correlation time related to ${}^3\text{He}$ diffusive motion, considering that T_2 is always much shorter than T_1 in low-dimensional systems [32,33]. They did not depend on n_3 , which implies that relaxations are determined by a single-particle process via diffusion in the local magnetic field such as by impurities, similar to high-temperature results observed in 1D channels of MCM-41 [21]. As a difference, the decreasing behavior of T_1 with T contrasts with T_1 increases observed in MCM-41 [21], which probably implies higher ${}^3\text{He}$ mobility in our condition using a FSM.

In the genuine 1D state of ${}^3\text{He}$ below 0.3 K ($\sim \Delta_{01}/2k_B$), several characteristic behaviors are observed as follows. As seen in Fig. 2, n_3 dependences of both T_1 and T_2 are developed below 0.3 K, which is consistent with the 1D state where collective motions among ${}^3\text{He}$ become dominant. In addition, transverse relaxation curves in this region fit the stretched exponential function $\exp[-(t/T_2')^\alpha]$ rather than the simple exponential function, where the exponent α starts to deviate below 0.3 K, from 1 at high T to about 0.7 at the lowest temperatures [28]. The stretched-exponential curves suggest a distribution of T_2 below 0.3 K, which is reasonable for 1D systems with large anisotropy of T_2 since the observed is a sum of signals from ${}^3\text{He}$ in many FSM grains with various orienta-

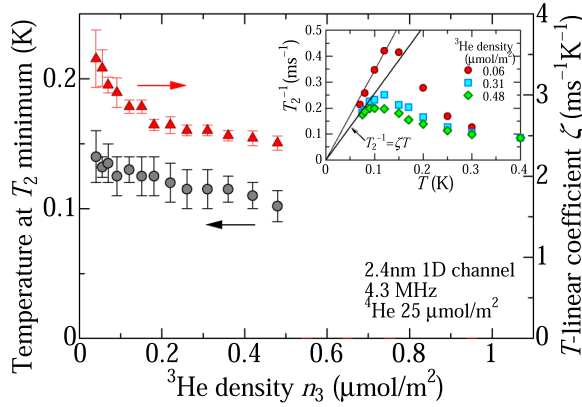


FIG. 3. Temperatures at the minimum of T_2 shown in Fig. 2(b) and proportional coefficients of the T -linear relaxation rate T_2^{-1} derived from linear plots of T_2^{-1} shown in the inset.

tions against the field. The most notable behaviors observed in the 1D region are upturns of T_2 below about 0.15 K, as shown in Fig. 2(b). In ^3He systems adsorbed on the wall, T_2 is often determined by motional narrowing due to diffusive motion of ^3He atoms. Therefore, T_2 monotonically decreases with T by suppression of thermally excited motion and becomes constant in the tunneling region at low T [32]. The behavior can be seen for T_2 of ^3He adsorbed in 3D interconnected nanopores ($T_F \approx 0.18$ K), shown in Fig. 2(b) as an example of other ^3He systems. Indeed, the approach to constant T_2 at low T has been observed also in these 1D channels at $n_3 > 0.6 \mu\text{mol}/\text{m}^2 \gtrsim n_{\text{lim}}$, where azimuthal motional states are partially occupied even at $T = 0$. Thus, the T_2 increases at low T are likely to be a characteristic property of the genuine 1D system. As shown in Fig. 2(b), T_2 increases are inversely proportional to the temperature; that is, relaxation rates T_2^{-1} are linear in T . The characteristic parameters, temperatures at the T_2 minimum and proportional coefficients at low T , are summarized in Fig. 3.

Since ^3He atoms and their nuclear spins form a 1D fluid in this system, the ^3He NMR corresponds to an electron spin resonance (ESR) in electronic systems. In 1D spin chains, although isotropic exchange interaction J does not result in any linewidths corresponding to T_2^{-1} , a small anisotropic interaction δ causes T -linear linewidths η at $T < J/k_B$, written as $\eta_{\parallel} \sim 4\pi^3(\delta/J)^2(k_B/\hbar)T$ for anisotropy with the axis parallel to the applied field and $\eta_{\perp} = \eta_{\parallel}/2$ for anisotropy perpendicular to the field [34]. A similar behavior was also shown by an analysis of the spin sector of the TL liquid model [35]. Thus, T -linear ESR linewidths have been considered to be a typical signature of a TL liquid for electrons. In this 1D ^3He , the dipole-dipole interaction is anisotropic, so that similar behavior is expected. On the other hand, when the typical dipole-dipole energy δ in 1D channels is consid-

ered to be $(\mu_0/4\pi)(\gamma^2\hbar^2/4d^3)$ for an interatomic distance $d \sim 1$ nm, δ is estimated to be only 0.8 nK, which is too small compared with $J \sim \epsilon_F$ to yield T_2 on the order of milliseconds. The mismatch implies the relevance of the other interactions, such as atomic exchange in channels or edge effects due to a finite length of about 300 nm [19]. What is puzzling is that similar T_2 increases were also observed even in the nondegenerate region above T_F . For dilute ^3He below $0.2 \mu\text{mol}/\text{m}^2$, T_F is considered to be lower than 0.1 K. A model calculation by Yao and Oshikawa considering the dipole-dipole interaction for a ^3He fluid adsorbed in the realistic nanochannel geometry confirmed that relaxations similar to ESR of 1D systems appear in NMR of the ^3He fluid even without a lattice but also showed that T -linear T_2^{-1} will be observed only below T_F , together with estimation of T_2 a few orders longer than observed [36]. As a clue for the solution, it is noted that T_1 shown in Fig. 2(a) and T -linear coefficients of T_2^{-1} in Fig. 3 indicate qualitative differences below about $n_3 \sim 0.2 \mu\text{mol}/\text{m}^2$, which suggests that different relaxation mechanisms work in the dilute nondegenerate region. In the dilute region, T_1 shows a T dependence similar to T_2 , which can be considered just a property of the dipolar relaxation with rapid diffusive motion at $\omega\tau < 1$ [32]. If that is the case, ^3He diffusive motion is implied to be faster with decreasing T , which is a quite characteristic behavior of this 1D system. Thus, the direct comparison with the TL liquid picture still leaves several issues. Nevertheless, the fact that T -linear T_2^{-1} are observed only in the 1D state indicates that this behavior is a reflection of the one dimensionality. The origin will be clarified by further studies under conditions with a different space, mobility, and interparticle interaction for ^3He , which can be modulated by the channel size and ^4He coating [19], together with the frequency dependences.

In conclusion, we have performed systematic NMR measurements for a quantum-mechanically 1D ^3He fluid formed in nanochannels, which is a candidate for the realization of a Tomonaga-Luttinger liquid. We found increases of the spin-spin relaxation time inversely proportional to T below 0.15 K, which is apparently similar to T -linear ESR linewidths for a spin TL liquid. While the origin is still controversial since similar behavior seems to be observed even in the nondegenerate region, it has been confirmed to be a characteristic behavior indicating the one dimensionality, which is observed only when the form, density, and temperature fulfill the 1D conditions for a ^3He fluid. Further experimental and theoretical studies are desired to reveal the 1D physics.

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