Relaxation of spin polarized ^{3}He in mixtures of ^{3}He and ^{4}He below the ^{4}He lambda point

Q. Ye, D. Dutta, H. Gao, K. Kramer, X. Qian, and X. Zong
Triangle Universities Nuclear Laboratory and Department of Physics, Duke University, Durham, North Carolina 27708, USA

L. Hannelius, R. D. McKeown, B. Heyburn, and S. Singer
Kellogg Radiation Laboratory and Department of Physics, California Institute of Technology, Pasadena, California 91125, USA

R. Golub
Department of Physics, North Carolina State University, Raleigh, North Carolina 27695, USA

E. Korobkina
Department of Nuclear Engineering, North Carolina State University, Raleigh, North Carolina 27695, USA
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We report a first study of the depolarization behavior of spin polarized ^{3}He in a mixture of ^{3}He-^{4}He at a temperature below the ^{4}He λ point in a deuterated tetraphenyl butadiene-doped deuterated polystyrene (dTPB-dPS) coated acrylic cell. In our experiment the measured ^{3}He relaxation time is due to the convolution of the ^{3}He longitudinal relaxation time, T_1, and the diffusion time constant of ^{3}He in superfluid ^{4}He since depolarization takes place on the walls. We have obtained a ^{3}He relaxation time of ~3000 s at a temperature around 1.9 K. We have shown that it is possible to achieve values of wall depolarization probability on the order of $(1-2)\times 10^{-7}$ for polarized ^{3}He in the superfluid ^{4}He from a dTPB-dPS coated acrylic surface.

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Hyperpolarized ^{3}He based on the technique of optical pumping [1,2] has found applications in diverse fields such as in the study of quantum phenomena in low-temperature fluids [3] and in the search for violations of fundamental symmetries [4–6]. They are also routinely used as polarized neutron spin filters [7], as effective polarized neutron targets for nuclear and particle physics experiments [8] and for low magnetic field magnetic resonance imaging [9]. All such applications have motivated, as well as benefited from, studies of the relaxation mechanisms of polarized ^{3}He in gas, liquid, and superfluid phases and under different surface conditions. The study of relaxation of polarized ^{3}He in ^{3}He-^{4}He mixtures at low temperatures is, however, of longstanding interest in its own right [10]. The simple atomic structure of ^{3}He makes a system of ^{3}He atoms ideal to model correlated fermions.

Superthermal production of ultracold neutrons (UCN) from superfluid ^{4}He has been demonstrated [11] as an efficient way of producing a large number of UCNs. The capability of storing a large number of UCNs following their production is important for experiments studying fundamental properties of the neutron, for example, the experiment on the search of the neutron electric dipole moment [6]. In this experiment, the deuterated tetraphenyl butadiene-doped deuterated polystyrene (dTPB-dPS) coated acrylic surface is chosen for such an application because of the small neutron absorption rate on the surface and its wavelength shifting property. The focus of this work is a first study of polarized ^{3}He relaxation time in a mixture of ^{3}He-^{4}He below the ^{4}He λ point in a dTPB-dPS coated acrylic cell. Such a study may find applications in the development of cryogenic ^{3}He magnetometers for experiments where trapping of polarized UCNs is involved as well as in other types of applications where polarized ^{3}He atoms are employed at low temperatures. At present the feasibility of ^{3}He magnetometers for UCNs has been studied only at room temperature [12].

While a number of experiments [13–17] have reported ^{3}He longitudinal relaxation times (T_1) in mixtures of ^{3}He-^{4}He at temperatures similar to our work, the measurements most relevant to ours are [14,16,17]. It was observed that ^{3}He atoms in a gaseous phase [16] in the presence of ^{4}He had a longer T_1 below temperatures where superfluid ^{4}He film was formed. Lowe *et al.* [14] observed little ^{3}He concentration dependence in the observed ^{3}He T_1, which was shorter than 300 s in ^{3}He-^{4}He solutions between 1.5 and 3.3 K. In this paper we report the first results of the ^{3}He relaxation time[1] in the presence of superfluid ^{4}He film and liquid in a dTPB-dPS coated acrylic cell at a temperature of 1.9 K and at a magnetic field of 21 G.

We have adopted the spin-exchange optical pumping (SEOP) technique for producing polarized ^{3}He nuclei. The polarization is measured using the adiabatic fast passage (AFP) technique of nuclear magnetic resonance (NMR). The schematic of the entire apparatus is shown in Fig. 1. It consists of a pair of Helmholtz coils with a diameter of 68 in. and the typical magnetic holding field is 21 G. A two-chamber apparatus for polarizing ^{3}He nuclei and for measuring their relaxation time at cryogenic temperatures is constructed from aluminosilicate glass (GE180), and a cylindrical acrylic cell. The two chambers are connected via a 3-mm-diameter, 21-in.-long pyrex capillary tubing, and are separated by a glass valve. The top chamber is a spherical cell with a diameter of 2.0 in., while the bottom chamber is a cylindrical acrylic cell with an outer (inner) diameter of

[1]The measured ^{3}He relaxation time is due to the convolution of the ^{3}He longitudinal relaxation time, T_1, and the diffusion time constant of ^{3}He in superfluid ^{4}He.

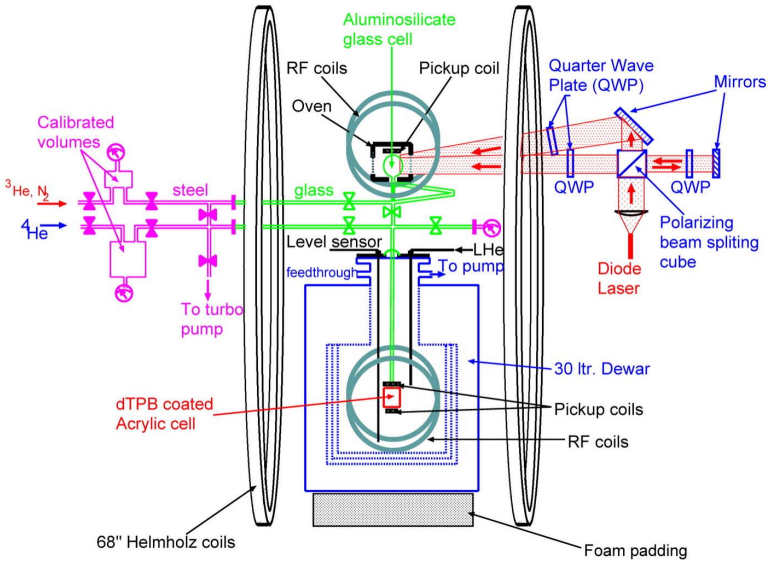

FIG. 1. (Color online) A schematic of the experimental setup.

2.0 in. (1.45 in.) and a length of 2.0 in. attached to the glass via a 0.5-in.-long glass to copper seal with a diameter of 3 mm. The copper seal is attached to the acrylic cylinder using the low-temperature epoxy Emerson and Cuming Stycast 1266. The inner surface of the acrylic cell is coated with dTPB-dPS. Details of the coating procedure can be found in [18]. Each chamber can be independently evacuated and filled with either ^{3}He and nitrogen gas[2] (top chamber) or ^{4}He gas (bottom chamber), and they can be isolated from the gas handling system via a pair of glass valves. The temperature of the bottom cell can be lowered to ~1.8 K by pumping on the vapor above the liquid helium inside the Dewar. The temperature is measured using a calibrated cernox resistance thermometer.

Two NMR systems are built in order to measure the ^{3}He relaxation time in both the top (pumping) cell and the bottom cell. Each NMR setup consists of a pair of RF coils, 12 in. in diameter and one or two pickup coils[3] with a diameter of 1.5 in. The RF and pickup coils for the low temperature (bottom) cell are placed inside the Dewar and hence are immersed in liquid helium during the measurement cycle.

The top cell is prepared by baking it under vacuum at ~350 °C, and then distilling a few milligrams of Rb into the cell (also under vacuum). Once the top cell is ready, a known amount of ^{3}He is introduced into the cell for each measurement (the amount can be varied as desired) and N_2 (~50–100 torr filled at room temperature) is also added as a buffer gas. The top cell is enclosed in an oven and heated to 190 °C and a 30 W circularly polarized laser light at 794.7 nm is incident onto the cell to polarize the ^{3}He atoms through SEOP. While the ^{3}He atoms in the top cell are being polarized, liquid ^{4}He is filled into the Dewar and the temperature of the bottom (acrylic) cell is lowered below the liquid ^{4}He boiling temperature by pumping on the ^{4}He vapor with a large throughput pump. Once the acrylic cell has reached the desired temperature with the lowest temperature being 1.8 K, a known amount of ^{4}He gas is introduced into the acrylic cell. The laser is then turned off and the top cell is cooled to room temperature, after which the glass valve separating the two chambers is opened to allow the polarized ^{3}He atoms to diffuse to the bottom acrylic cell. The N_2 gas condenses on the way down and does not enter the bottom cell. The valve is closed after 30 s and a series of NMR-AFP measurements are performed with a time interval between 50–220 s. The amount of ^{3}He in the capillary tube in our experiment is negligible.

Measurements are carried out with a dTPB-dPS coated acrylic cell. The relaxation time of ^{3}He is consistently shorter than 10 s with no ^{4}He inside the cell at a temperature of around 1.9 K. A strong correlation between the ^{3}He relaxation time and the amount of ^{4}He atoms introduced into the cell is observed. Further, relaxation times in excess of 3000 s are observed. For comparison, the ^{3}He relaxation times at room temperature from the optical pumping GE180 glass cell are between 5980 and 6700 s. Figure 2 shows the ^{3}He relaxation times at 21 G holding field from a dTPB-dPS coated acrylic cell at ~1.9 K. The amount of ^{4}He is varied from 0.0 to 1.076 mol while the amount of ^{3}He is fixed at 0.0014 mol.[4] ^{3}He relaxation times are extracted by fitting the NMR data as a function of time to an exponential decay form with corrections for AFP spin-flipping inefficiency. The AFP spin-flipping inefficiency is determined to be $(1 \pm 1)\%$. The longest ^{3}He relaxation time obtained at ~1.9 K from the dTPB-dPS coated acrylic cell is 3152 ± 86 (statistical) ± 473 (systematic) seconds. The main contributions to ^{3}He depolarization are the dipole-dipole relaxation mechanism, the magnetic field gradient effect and the surface effect at the wall. For the data shown in Fig. 2 the dipole-dipole relaxation time is calculated to be 6.24×10^5 s [19]. The relaxation time due to the magnetic field gradient in our system is

[2] N_2 gas, introduced for efficient optical pumping, will freeze on the wall of the capillary tube at low temperatures.

[3] Two pickup coils are attached to the dTPB-dPS coated acrylic cell as shown in Fig. 1.

[4] The temperature range for these measurements is from 1.83 to 1.90 K.

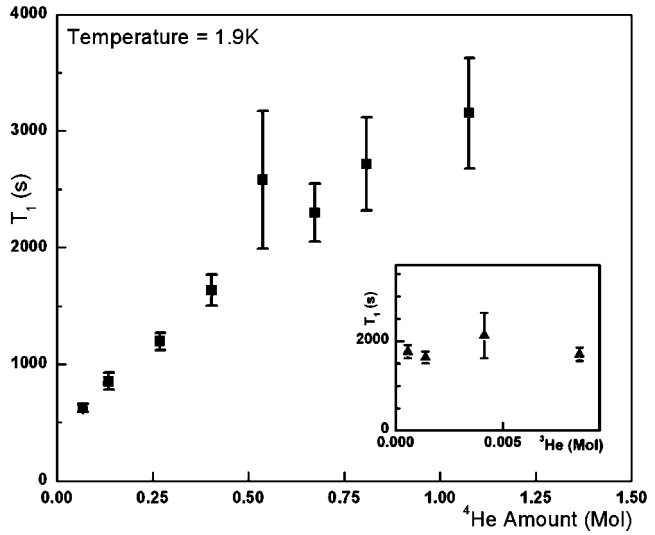

FIG. 2. The relaxation time of polarized ^{3}He as a function of the amount of ^{4}He in the measurement cell at a temperature of ⟨1.9 K⟩. The error bars are the quadrature sum of the statistical and systematic uncertainties. The figure inset shows the polarized ^{3}He relaxation time as a function of the amount of ^{3}He, when the amount of ^{4}He is held constant at 0.404 mol.

studied carefully using a NIST GE180 sealed cell,[5] which is filled with 100 torr of ^{3}He, 50 torr of N_2, and 534 torr of ^{4}He, at room temperature. From these studies we extract the magnetic field gradient at 300 K at the position of the measurement cell. The ^{3}He relaxation time at 1.9 K due to this magnetic field gradient is calculated to be 5.26×10^5 s. Therefore, the surface effect at the walls is the most important contribution to the ^{3}He relaxation time in our measurements.

The initial improvement observed in the ^{3}He relaxation time shown in Fig. 2 can be attributed to the formation of a superfluid ^{4}He film on the dTPB-dPS coated acrylic wall. However, the thickness of this film varies extremely slowly with the amount of ^{4}He [20] for the entire range of our measurement. The behavior of the ^{3}He relaxation time as the amount of ^{4}He is increased, can be understood using simulations described below.[6]

The ^{4}He atoms liquefy and collect at the bottom of the cell with height ranging between 0.0675 mol (0.17cm) and 1.076 mol (2.71cm) as shown in Fig. 2. The approximate time required for diffusion over a given distance h is $\frac{h^2}{2D}$. The estimated diffusion time of ^{3}He from the top of the liquid surface to the bottom ranges between ~60 and 15 300 s as the height of the liquid increases. Our system was far from equilibrium for most of the measurements we took. A time-dependent diffusion model and a static magnetic field model are developed to simulate the signal in the pickup coil in our experiment.

The inset in Fig. 2 shows the measured ^{3}He relaxation time versus the amount of ^{3}He (in units of mole) in the cell for a fixed amount of (0.404 mol) ^{4}He. Our results show that the relaxation time (~1800 s) is almost independent of the amount of ^{3}He in the range of our measurement (0.000 56 −0.0086 mol). The model used to analyze the data assumes that all ^{3}He atoms are in the vapor state immediately after the ^{3}He atoms enter the acrylic cell. The concentration is assumed to be uniform in the vapor, and zero in liquid ^{4}He. ^{3}He atoms diffuse both in the vapor and liquid, in which the diffusion coefficients are different. At 1.9 K, $D_l=2.4\times10^{-4}$ cm^2/s [21] is the diffusion coefficient of ^{3}He in liquid ^{4}He. The vapor ^{3}He diffusion coefficient is calculated using $D_v=1.463\times10^{-3}T^{1.65}P^{-1}=0.018$ cm^2/s [22].

The boundary condition at the liquid surface is written using the flux exchange between the vapor and liquid. The flux going from vapor to liquid is $|\vec{j}_{vl}|=\frac{1}{4}v_v n_v$ and in the opposite direction $|\vec{j}_{lv}|=\frac{v_v}{4}(\frac{m}{m^*})^{3/2}e^{-E_B/kT}n_l$. n_v and n_l are the concentration of the polarized ^{3}He atoms in the vapor and liquid, respectively. The average speed of ^{3}He in the vapor is $v_v=\sqrt{\frac{8kT}{\pi m_3}}=1.15\times10^4$ cm/s. The effective mass of ^{3}He dissolved in superfliud ^{4}He is $m_3^*=2.4m_3$, where m_3 is the mass of a ^{3}He atom.

The pickup coil is mounted at the bottom of the acrylic cell and it measures the change of the magnetic flux caused by the spin-flip of the ^{3}He magnetic dipoles in the cell (both in the vapor and in the liquid) during an NMR-AFP sweep. In order to calculate this flux, we use the reciprocity theorem, according to which the flux through the pickup coil can be calculated as proportional to the field produced by a current in the pickup coil at the location of the ^{3}He dipole.

Our measurements are best characterized by the depolarization probability (DP) per wall collision. In the analysis we allow for this probability to be different on the walls covered with bulk liquid, P_l, and the walls covered with superfluid film only, P_v. The wall boundary condition is that the depolarization rate on the wall is the product of the number of atoms reaching the wall per unit time and the corresponding DP. The depolarization behavior of the model can be changed by varying these parameters. To extract these parameters we did least-squares fits of the measured ^{3}He NMR-AFP signal versus time from injection of the polarized ^{3}He into the ^{4}He containing cell, with the same quantity calculated by solving the diffusion equation as specified above and using the solutions to calculate the signal in the pickup coil.

Because ^{3}He atoms dissolve into the liquid ^{4}He rapidly without losing polarization, the signal increases from zero in the beginning of the measurement and then decays after it saturates. P_v will influence the short time buildup of the signal in the pickup coil and P_l will determine the long time behavior. Thus a larger P_v and smaller P_l, will move the peak of the signal to shorter times. The pickup coil is located on the bottom of the measurement cell and so is more sensitive to ^{3}He dissolved in the liquid. And it becomes less sensitive to the ^{3}He in the vapor as the amount of ^{4}He is increased. In the measurements made with small amounts of ^{4}He the long time behavior is influenced by both P_v and P_l. So it is difficult to extract unique values of the parameters from these data. As an example Figs. 3(a) and 3(b) show the varying contribution of the ^{3}He in the liquid (pink circles) and vapor

[5]The cell is on loan from Gentile at NIST.

[6]The COMSOL Multiphysics finite element package is used to solve the diffusion equations in our analysis.

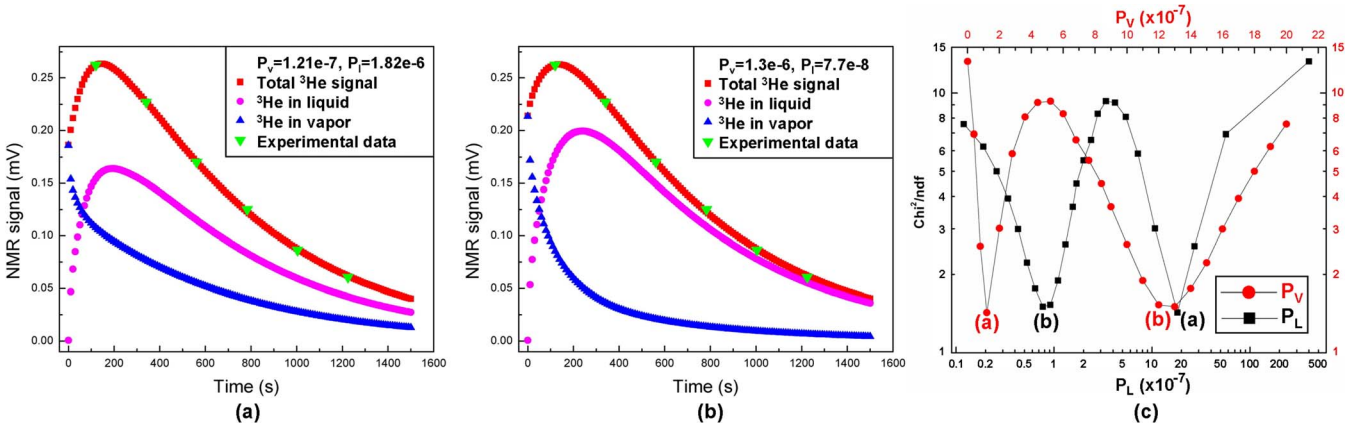

FIG. 3. (Color online) (a) and (b) are NMR measurements of the ^{3}He signal (green triangles) at 1.9 K as a function of time with the amount of ^{4}He equal to 0.135 mol (0.34 cm). Red squares are the simulated total signal in the pickup coil consisting of the contributions from the vapor (blue triangles) and liquid (pink circles). (c) is reduced χ^2 obtained from the best fit as a function of P_v (red circles, top axis) and P_l (black squares, bottom axis) showing how different values of P_v and P_l can fit the data due to the fact that with low liquid level the vapor is close to the pickup coil.

(blue triangles) to two equally good fits (red squares and green triangles) for $P_{v,l}$ varying by about a factor of 10. Figure 3(c) shows the plot of reduced χ^2 obtained from the best fits as a function of P_l (bottom axis) and P_v (top axis). For larger ^{4}He quantities the results are not sensitive enough to P_v to allow the extraction of a value for this quantity.

The fitting is made more difficult in that we have no absolute polarization information so that we must treat the normalization of the curves as a free parameter. This is another reason that a range of parameters can give good fits in the low filling cases. In addition, our operational procedures were such that in most cases we started taking data after the peak had been passed. Only the run with 0.673 mol (1.69 cm) of ^{4}He shows the peak of the signal (Fig. 4) and we are able to extract a reasonable value of P_l from the fit (Fig. 5), $P_l=(3.9^{+2.0}_{-0.7})\times10^{-7}$. The error bar is determined by the standard method of varying the P_l parameter so that the reduced χ^2 is increased by 1. For measurements with larger amounts of ^{4}He, diffusion to the walls plays a significant role and calculations show that the long time behavior is less sensitive to the value of P_l so we cannot extract meaningful values of the wall loss parameters from the data. In Fig. 5, we also show the reduced χ^2 plots for the runs with 0.404 mol (1.02 cm) and 0.538 mol (1.35 cm). From these plots we can extract $P_l=(1.7\pm0.2)\times10^{-7}$ and $P_l=(1.6\pm0.4)\times10^{-7}$, respectively. The minima in reduced χ^2 are much broader when plotted versus P_v so we cannot extract useful values for this parameter.

Lusher *et al.* [16] carried out a series of measurements with open Pyrex glass chambers as well as sealed Pyrex glass cells. Their results showed that the formation of a superfluid ^{4}He film on a hydrogen coated glass surface reduces the depolarization of ^{3}He from the surface. For an open cell they

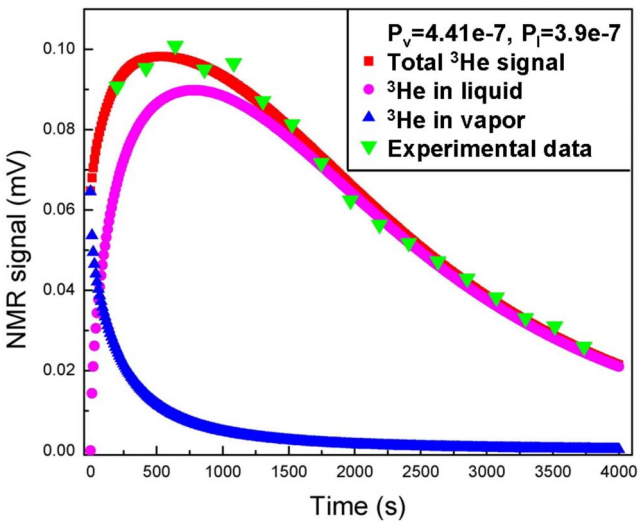

FIG. 4. (Color online) The amount of ^{4}He in the acrylic cell is 0.673 mol (1.69 cm). The experimental data (green triangles) consisting of the contributions from the vapor (blue triangles) and liquid (pink circles) are fitted onto the simulation results (red squares).

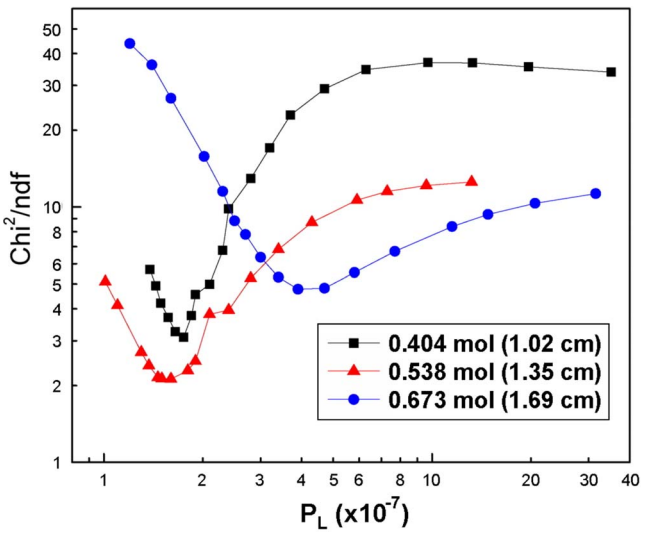

FIG. 5. (Color online) Reduced χ^2 for the fits versus P_l values for ^{4}He amounts of 0.404 mol (1.02 cm), 0.538 mol (1.35 cm), and 0.673 mol (1.69 cm).

observed a relaxation time of ~500 s at a magnetic holding field of 0.23 tesla and a temperature of 1.9 K. The ^{3}He bulk number density for these measurements was 5.2×10^{-6} mol/cc (cell volume 4.2 cc) and the ^{3}He:^{4}He atomic ratio was 1:16 (ours is 1:769). As shown in Fig. 2 we have observed relaxation times in excess of 3000 s at 1.9 K for a holding field of 21 G. The surface-to-volume ratio of our cell is 50% of the cells used in measurements of [16], and our measured relaxation time is a convolution of ^{3}He T_1 and the ^{3}He diffusion time constant. Their corresponding depolarization probability is determined to be $\sim 1.9 \times 10^{-7}$, which is similar to our P_l value, though ours is obtained from a dTPB-dPS coated acrylic surface under the superfluid ^{4}He liquid.

We have measured the relaxation time of polarized ^{3}He in a dTPB-dPS coated acrylic cell in a diluted mixture of ^{3}He-^{4}He at a temperature of 1.9 K with a magnetic holding field of 21 G. We have shown that it is possible to achieve values of wall depolarization probability (P_l) on the order of $(1-2) \times 10^{-7}$ for polarized ^{3}He in the superfluid ^{4}He at 1.9 K. To provide precise determination of these depolarization probabilities in future measurements, one needs to isolate the diffusion time scale from the system, i.e., to carry out measurements in a cell with superfluid ^{4}He film on the wall only, and measurements from a cell filled with superfluid ^{4}He completely. It also remains to be seen how sensitive depolarization probabilities are to surface preparations. Nevertheless, ours is the first study of the polarized ^{3}He relaxation time from dTPB-dPS coated surfaces in superfluid ^{4}He. Our data suggest that such surface may find applications in areas which employ polarized ^{3}He at low temperatures in the environment of superfluid ^{4}He. Since the ^{3}He behavior is mostly dominated by diffusion in liquid ^{4}He at 1.9 K, it is important to extend our current work to below 1 K due to much shorter ^{3}He diffusion time. Such measurements are currently in progress.

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