

^3He Spin Diffusion Measurements in ^3He - ^4He Mixture Films

P. A. Sheldon* and R. B. Hallock

Laboratory for Low Temperature Physics, Department of Physics, University of Massachusetts at Amherst, Amherst, Massachusetts 01003

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^3He spin diffusion measurements for ^3He - ^4He mixture films on Nuclepore are reported as a function of ^3He coverage for $0.030 \leq T \leq 0.070$ K. We report measurements for a constant ^4He coverage of 5.0 bulk density atomic layers, with ^3He coverages between 0.10 and 0.98 layers. We find that the ^3He spin diffusion coefficient, as measured by Hahn spin echoes, has an unexplained maximum as a function of ^3He coverage and that the temperature dependence is somewhat weaker than predicted. Comparison measurements are presented for the NMR relaxation times T_1 and T_2 and the magnetization over the same ^3He coverage range.

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Helium adsorbs uniformly to most substances, and at low temperatures, for ^4He films thicker than a few atomic layers, the helium is a superfluid. When atoms of ^3He are added to this system at low temperatures, they reside on the free surface of the ^4He film, which is an almost ideal two-dimensional environment. The study of such a mixture film system provides great insight into the physics of uncharged Fermi particles in two dimensions. NMR is a remarkably effective tool for the study of the ^3He in such ^3He - ^4He mixture films. Previous work [1] has established that for $T \leq 250$ mK submonolayer amounts of ^3He adsorbed to a ^4He film reside in a ground state on the free surface of the ^4He film and behave as a two-dimensional Fermi gas [2,3]. An increase in the amount of ^3He in the film causes an increase in the Fermi temperature and an increase in the interactions among the ^3He , but at low enough temperatures the system remains a well-behaved two-dimensional degenerate Fermi gas. Excited states for the ^3He also exist in the ^4He film and these can be populated by an increase in the temperature or by an increase in the ^3He coverage [4,5]. The motion of the ground-state ^3He atoms in the plane of the ^4He film also shows interesting behavior. Measurements of the ^3He spin diffusion have shown evidence of localization for small amounts of ^3He on very thin ^4He films. To observe this localization, the ^4He film thickness on which the ^3He resides was varied. As the ^4He film thickness is increased from 2 to 3 atomic layers, a sharp rise is observed in the diffusion coefficient, D , indicating the delocalization of the ^3He atoms with increasing ^4He film thickness [6–9]. For ^4He coverages above 4 atomic layers D depends weakly on ^4He coverage. As a complement to this earlier work we have measured and report here the ^3He spin diffusion coefficient as the ^3He coverage is increased for fixed ^4He coverage. Such changes in ^3He coverage cause the ^3He - ^3He interactions to increase, and this is expected to influence the spin diffusion. We employ a ^4He coverage of 5.0 bulk-density layers, thick enough to be a superfluid [10] and well above the ^4He coverage range [7,9] at which localization of the ^3He is observed.

We used pulsed NMR techniques at 62.9 MHz in a 2 T field to make measurements of the longitudinal spin relaxation time, T_1 , the transverse spin relaxation time, T_2 , the magnetization, M , and the spin diffusion coefficient, D , of the ^3He atoms that are adsorbed on the ^4He film. In this Letter we concentrate on the spin diffusion measurements, but we also report data for the behavior of T_1 , T_2 , and the magnetization for comparison. To determine the diffusion coefficient, Hahn spin echos [11] were measured in a linear gradient field ($0 < G < 10$ G cm $^{-1}$). Fits to the pulse echo heights E as a function of G provide us with the transverse diffusion coefficient D by use of

$$E(\tau, G) = M_0 \exp\left[-\frac{2\tau}{T_2} - \frac{2}{3} \gamma^2 D G^2 \tau^3\right], \quad (1)$$

where M_0 is the magnetization, γ is the gyromagnetic ratio, and τ is the time between $\pi/2$ and π pulses. The pulse echo height is measured for several values of τ and G , and the slopes of $\ln(E)$ vs τ are determined for each value G . The resulting slopes' linear dependence on G^2 are used to determine D .

The helium films are adsorbed onto a Nuclepore [12] substrate, a polycarbonate sheet 10 μm thick perforated with 2000 \AA diameter pores at a density of $\sim 3 \times 10^8$ pores cm $^{-2}$. For the coverages used in this work, helium does not capillary condense on this substrate [13], ensuring a two-dimensional substrate for the ^3He . Since the ^3He diffusion during a Hahn spin echo pulse sequence may involve transit distances on the order of μm , this Nuclepore surface hinders the diffusion as the ^3He atoms move along and among the pores. The measured diffusion coefficient D is therefore proportional to the diffusion coefficient \mathcal{D} that would be expected were it not for the tortuosity of the substrate. The proportionality constant due to this tortuosity for Nuclepore was determined theoretically [14] to be $\phi = 16$ and was previously measured macroscopically by third sound techniques by Smith *et al.* [15] and found to be $\phi \approx 14$. For Hahn spin echos in this context, it is estimated [7] that for the finite diffusion distances encountered here $\phi_{\text{effective}} \approx 0.8\phi = \phi_e$; thus, for

our purposes $\mathcal{D} = \phi_e D$. All of the data to be presented here is for the measured diffusion coefficient D .

The experimental cell [16] used for these measurements is the same as that used in references [2–5]. It has a volume of 52 cm³ and a total surface area of 1.77 cm². The substrate for the NMR measurements consists of a stack of 393 Nuclepore disks inside the NMR coil. The disks are oriented with the plane of the face of the disks parallel to the polarizing and gradient fields but perpendicular to the NMR RF field; the normal to the plane of the disks is parallel to the NMR RF field. The cell is mounted to the mixing chamber of a dilution refrigerator with each Nuclepore disk in radial compression contact with a copper post inside the sample cell [16].

Measured amounts of ³He and ⁴He gas at room temperature were added to the sample chamber at $T \leq 150$ mK to form the films under study. Beginning with 5.0 layers of ⁴He we made incremental additions of ³He beginning with 0.1 layer of ³He. Following each addition of ³He, the sample was annealed at $T \geq 500$ mK to ensure mixing. The various measurements were then made as a function of temperature, after which the next increment of ³He was added to the cell. By our definition, a coverage of one atomic layer corresponds to the number of atoms necessary to cover the entire surface area with one monolayer at bulk-liquid density. One layer corresponds to 0.064 ³He atoms Å⁻² or 0.077 ⁴He atoms Å⁻². Of course, the first ⁴He monolayer adsorbed achieves a density significantly higher than the bulk density.

At a coverage of 5.0 bulk-density layers of ⁴He we made incremental additions of ³He from 0.10 layers up to 0.98 layers and studied D at several temperatures. Here to focus the discussion we confine attention to the data for $T < 100$ mK. Data for several fixed temperatures as a function of ³He coverage reveal significant coverage-dependent structure which sharpens with decreasing temperature as shown in Fig. 1. At a ³He coverage of 0.1 layers, our present data is generally consistent with earlier data [6–9]. As the ³He coverage is increased, D increases with coverage at low coverage; then it plateaus until the coverage reaches ~ 0.6 layers. The increase in D with coverage at low coverages is especially strong for the lowest temperatures studied. This increase is surprising and we do not understand it. One might expect that as the ³He coverage is increased interactions and scattering would increase and this would lead to a reduction in the diffusion coefficient. On the other hand, an increase in ³He coverage raises T_F and decreases the population available for scattering at a given temperature, which tends to increase D .

A careful theoretical analysis of the spin diffusion of a Fermi liquid in two dimensions has been carried out by Miyake and Mullin [17] with the result that for our experimental conditions

$$D = (h/2\pi^2 m^*) (1 + F_0^A)^3 (T_F/T)^2 \times C(\lambda_-) / [(F_0^A)^2 \ln(T_F/T)], \quad (2)$$

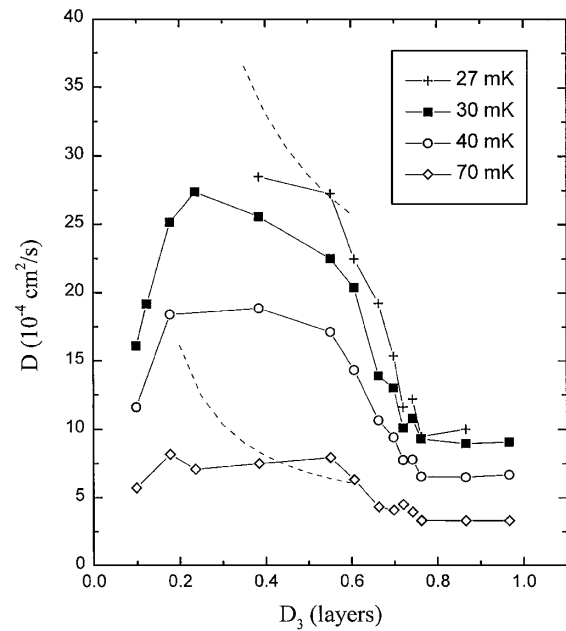


FIG. 1. The diffusion coefficient D versus ³He coverage in bulk-density atomic layers. Temperatures are indicated in the upper right in mK. The lines connecting the symbols are guides to the eye. The dashed curves are theoretical predictions based on Eq. (2) and adjusted for the tortuosity by use of $\phi_e = 12$.

where F_0^A is a Fermi liquid parameter, h is Planck's constant, $C(\lambda_-)$ is a numerical factor between 3/4 and 1, and the effective mass is given by $m^* = m_h(1 + F_1^S/2)$ with m_h the hydrodynamic mass. For the conditions of our experiment we expect $C(\lambda_-) \approx 3/4$. Equation (2) allows us to predict the coverage and temperature dependence of D . An evaluation of Eq. (2) for D as a function of ³He coverage requires data for F_0^A and F_1^S . Using recent results for F_1^S for ³He-⁴He mixture films on graphite [5] and on Nuclepore [18], and results for $M \sim (1 + F_1^S/2)/(1 + F_0^A)$ on Nuclepore to determine F_0^A as a function of ³He coverage, Eq. (2) predicts D to be a decreasing function of ³He coverage over the coverage range studied here. We observe D to increase with coverage at low coverages, which remains unexplained. For $D_3 > 0.3$ layers, the magnitude of D is reasonably close to the predicted values. For example, the two dashed curves shown on Fig. 1 represent Eq. (2) for $0.2 \leq D_3 \leq 0.6$ layers, with $C(\lambda_-) = 3/4$ and tortuosity $\phi_e = 12$ for the two cases $T = 30$ mK and 70 mK.

Concurrent measurements of the NMR's T_1 and T_2 and the magnetization (Fig. 2) show that the relaxation times have weak coverage dependence for $0.1 < D_3 < 0.6$ layers. As the ³He coverage is increased, promotion to the first excited state takes place as evidenced by the step in the magnetization [4] in the vicinity of 0.65 ³He layers. The NMR's T_1 and T_2 also show some structure in the vicinity of the step in the magnetization as shown in Fig. 2. The strong decrease in the diffusion coefficient in the vicinity of promotion of ³He to the first excited state is no doubt related to the population of this excited state. With

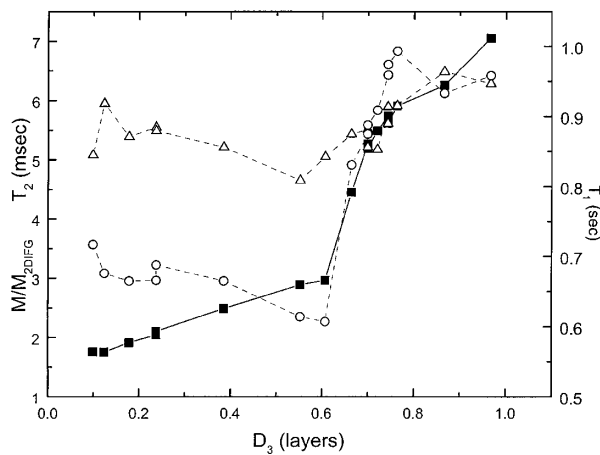


FIG. 2. The NMR T_1 (circles), T_2 (triangles), and magnetization $M/M_{2\text{DIFG}}$ (solid squares) versus ^3He coverage for $T = 30$ mK, where $M_{2\text{DIFG}}$ is the magnetization of an ideal two-dimensional Fermi gas. The solid line and dashed lines connecting the data points are guides to the eye. No low-coverage structure reminiscent of that seen in D is present in these quantities. The decrease in D near 0.65 ^3He layers appears related to the occupation of the first ^3He excited state as evidenced by the step [4] in the magnetization.

such promotion some of the ^3He are believed to be deeper within the ^4He film [19] and this perhaps presents a more constrained environment leading to more scattering and an enhanced reduction of the diffusion coefficient. ^3He coverages above 0.8 layer show a nearly constant diffusion coefficient.

D exhibits a modest evolution in temperature dependence with ^3He coverage (Fig. 3), but the observed temperature dependence is somewhat weaker than that predicted by Miyake and Mullin [17] for a weakly polarized two-dimensional degenerate Fermi gas: $\mathcal{D} \propto 1/[T^2 \ln(T_F/T)]$. The data shown in Fig. 3 are below the degeneracy temperature and show a decreasing diffusion coefficient with increasing temperature, consistent with earlier work, but the data are not fully consistent with the temperature dependence predicted by theory. For low ^3He coverages on similar ^4He films, Sprague *et al.* [6,7] also observed a spin diffusion coefficient with a temperature dependence weaker than T^{-2} . The solid line in Fig. 3 has the temperature dependence $D \sim A/[T^2 \ln(T_F/T)]$ and is shown with an arbitrary choice for the coefficient A . Although the temperature range is relatively narrow for meaningful power law fits, fits to the data at different ^3He coverages of the form $D \propto T^{-p}$ were carried out to characterize the data, and two representative fits are shown in the figure by dashed lines. The results for the exponent p are shown in the lower inset as a function of the ^3He coverage. It is observed that $p \sim 1.3$, with p weakly decreasing with increasing ^3He coverage.

In summary, we have observed a significant and not fully understood peak in the diffusion coefficient as a function of submonolayer ^3He coverage for degenerate two-

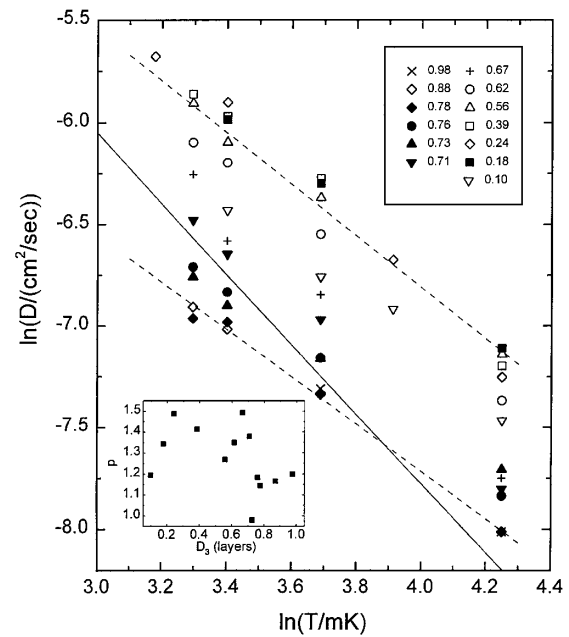


FIG. 3. The diffusion coefficient $\ln(D)$ versus $\ln(T)$. Coverage values in bulk-density ^3He atomic layers are indicated in the inset at the upper right. The solid line has the temperature dependence of Eq. (2). The dashed lines are fits to the data of the form $D \sim T^{-p}$ for two representative cases, $D_3 = 0.56$ (upper) and 0.88 (lower) layers. The inset at the lower left shows the exponent p as a function of ^3He coverage.

dimensional ^3He films that reside on a ^4He film at low temperatures. Otherwise, the measured diffusion coefficient is in reasonable accord with theoretical predictions, although the variation of D as a function of temperature is somewhat weaker than expected for the full range of ^3He coverages studied here.

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*Present address: Randolph-Macon Woman's College, Lynchburg, VA 24503

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