^{3}He Mobility and Localization in Thin ^{4}He Films

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We report measurements of spin-diffusion coefficients for 0.1 monolayer of ^{3}He adsorbed on thin ^{4}He films as a function of temperature and ^{4}He coverage. For ^{4}He coverages below a critical coverage D_c the ^{3}He are immobile. The ^{3}He quasiparticles in the film experience a mobility edge as the ^{4}He coverage is increased beyond D_c. This is accompanied by a vanishing of local moments as evidenced by the disappearance of a Curie-like component of the magnetization for coverages $D > D_c$. A peak in the transverse relaxation T_2 occurs near D_c.

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In thin adsorbed films of ^{3}He-^{4}He mixtures, low coverage ^{3}He is appropriately described at low temperatures ($T < 200$ mK) as a two-dimensional weakly interacting Fermi gas [1,2] occupying the lowest energy bound state at ^{4}He film free surface. The energetics and thermodynamic properties of the bound states are sensitive to the structure of the liquid-vapor interface of the film [3,4] and exhibit [1,5–7] a dependence on the ^{4}He areal coverage. Transport properties are similarly affected by the local ^{4}He density and Fermi interactions [8,9]. Theoretical treatments [3,4] invoke translational invariance of the ^{3}He environment and have been quite successful in predicting the ^{3}He effective mass and the energies of the ^{3}He states for ^{4}He films thick enough to be fluid. In contrast, the effect of translational disorder on the behavior of the ^{3}He impurities is not yet well known. Very thin ^{4}He films are expected to be highly disordered through local variations in binding potential and solid-layer roughness, and so for these rather different thin films translational invariance breaks down. The effective mass and the density of states for the ^{3}He impurities are sensitive to the potential structure, and hence spatial fluctuations in this potential may be expected to be a strong source of scattering. The ^{4}He film with ^{3}He impurities is analogous to two-dimensional electron systems, such as thin metallic films with quenched disorder, and to heterostructures, where theories for percolation and weak localization are well developed. Standard metal-insulator transition models consider quasiparticles moving in a randomly fluctuating perturbing potential [10,11]. The present work is an effort to explore this analogy by a study of the properties of neutral Fermi particles as a function of *in situ* variation of the potential caused by changing the ^{4}He film thickness D_4.

For the thinnest ^{4}He films we imagine that the variations in an effective potential are sufficiently strong to localize the ^{3}He atoms. Quasiparticles at the Fermi surface do not have sufficient energy to travel macroscopic distances, and hence no ^{3}He diffusion is expected. As the ^{4}He coverage is increased the potential evolves and at a critical ^{4}He coverage D_c, percolation is expected. As the coverage is further increased the topography of the evolving potential surface determines the diffusion. Eventually the average quasiparticle energy exceeds all variations of the binding potential and the influence of the perturbing potential can be thought of as a weak source of scattering. The measurements we report here show the ^{3}He diffusion to be a strong function of ^{4}He coverage and validate this general picture.

Here we report measurements of the spin diffusion coefficient D of ^{3}He in the quasi-two-dimensional ground state together with measurements of the spin susceptibility χ and the longitudinal and transverse relaxation rates $1/T_1$ and $1/T_2$, respectively, using pulsed NMR techniques [12] at 62.9 MHz. Our measurements are carried out on the polycarbonate substrate Nuclepore. 400 Nuclepore filters each perforated with 3×10^8 2000 Å diam holes/cm^2 provide a total surface area of 1.77 m^2 inside the ~1 cm^3 NMR coil. The susceptibility and relaxation measurements are made by the use of Hahn spin echoes and the inversion recovery technique, while stimulated echoes and spin echoes in a static field gradient g, $0 \leq g < 15$ G/cm, are used to measure the diffusion. The primary advantage of using stimulated echoes is that the decay of the echo amplitudes is not limited by T_2 (as with Hahn echoes) but by T_1, which can be quite long in this system ($1 \leq T_1 \leq 100$ sec). A submonolayer coverage of ^{3}He, $n_3 = 1.08$ μmoles/m^2 ($\approx$0.1 monolayer), is adsorbed on a thin ^{4}He film of varying thickness, $5.2 \geq D_4 \geq 2.1$ layers, on Nuclepore, where the ^{4}He coverage is $n_4 \equiv 12.82$ (μmoles/m^2)/layer $\times D_4$, with D_4 the thickness of the film expressed in terms of equivalent bulk-density layers.

During the course of a diffusion measurement, the ^{3}He may travel macroscopic distances (~μm), and so the large scale topography of the Nuclepore surface must be considered. A mean-field tortuosity factor α is defined by $D_m = D/\alpha$, where D_m is the measured diffusion coefficient with D the coefficient which would be present on a smooth surface. Third-sound velocity measurements can be used to determine α; we find [13,14] $\alpha \approx 16$. During a diffusion measurement which utilizes stimulated echoes, a ^{3}He atom visits many pores.

However, during the time span of a Hahn echo experiment ($T_2 \simeq 5$ ms), many spins do not have sufficient time to leave pores, and hence a different effective tortuosity can be expected. Comparison of diffusion data obtained by the two techniques suggests that $\alpha_{\mathrm{Hahn}} \sim 0.8\alpha$ is appropriate. We consider here the diffusion data derived primarily from the stimulated echo experiments.

In a constant field gradient g, a Hahn spin echo formed by a 90-τ-180 pulse sequence produces an echo of amplitude E at time 2τ, where $\ln(E) \propto -\gamma^2 g^2 \alpha t^{\kappa+2}$, while a pulsed field gradient spin echo or stimulated echo has an amplitude $\ln(E) \propto -\gamma^2 g^2 \delta^2 \Delta^{\kappa}$, where δ is the gradient pulse width, γ is the gyromagnetic ratio, $t = \tau$, and Δ is the gradient pulse separation or, for stimulated echoes formed by a 90-δ-90-Δ-90 pulse sequence in a constant field gradient, δ is the separation between the first and second rf pulses and Δ is the separation between the second and third pulses. κ is defined [15] by the time evolution of the mean-square displacement r in the external gradient $\langle r^2 \rangle \propto t^{\kappa}$. All of the diffusion data presented here are consistent with $\kappa = 1$, which implies that any anomalous diffusion is not due to percolated motion on a fractal network where $\kappa < 1$ might be expected.

For ^{4}He coverages $D_4 > 2.66 = D_c$, degenerate susceptibilities are observed with degeneracy temperatures T_F^{**} in the range $100 \leq T_F^{**} \leq 300$ mK, and the temperature dependence [7] is that of an ideal Fermi gas using a degeneracy temperature T_F^{**} in place of the Fermi temperature T_F [where $T_F^{**} = T_F^*(1 + F_0^a)$], with the T_F^{**} determined by fits to the data at each coverage and where T_F^* is the Fermi temperature replacing the ^{3}He mass by its effective mass m^*. However, for $0 \leq D_4 < D_c$ the magnetization contains a Curie component to the lowest temperatures investigated, $T \geq 24$ mK (see Fig. 1),

$$M = \frac{C}{T_F^{**}}\left(1 - e^{-T_F^{**}/T}\right) + \frac{C_0}{T}. \tag{1}$$

The Curie fraction $N_C/N = C_0/(C + C_0)$ is a function of the ^{4}He coverage and is associated with spins which we believe to be localized in the solid layer [16]. For coverages $D_4 \sim 2$ layers these ^{3}He account for $\sim$10% of the total ^{3}He coverage in the film [Fig. 2(a)].

The vanishing of the Curie fraction $N_C/N = C_0 = 0$ for $D_4 \geq D_c$ layers [see Fig. 2(a)] may be interpretated as the signal of liberation of localized ^{3}He from the solidified second layer of the ^{4}He film. Below this coverage, spin diffusion $D \sim 10^{-6}$ cm^2/sec is measured, and long ($\sim$10–100 sec) T_1 relaxation times are seen, both consistent with ^{3}He participating in a solidified second layer [Fig. 2(b)]. It is also apparent from the change in the temperature dependence of the T_1 data that there is a dramatic reduction in the spin-spin correlation τ_c time as we go from $\omega\tau_c \gg 1$ to $\omega\tau_c \ll 1$ as D_4 is increased beyond D_c layers. Note $T_1 \gg T_2$ [Figs. 2(b) and 2(c)], an observation [17] consistent with earlier measurements [18–20].

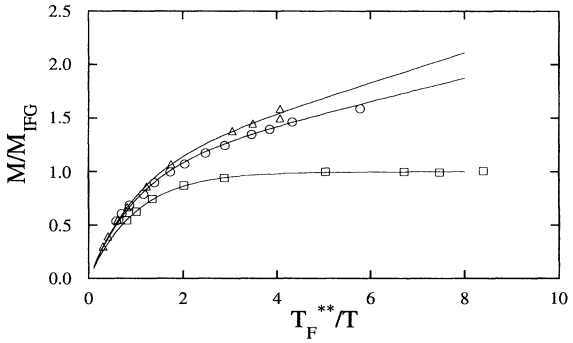

FIG. 1. Magnetization (normalized by the ideal two-dimensional ideal Fermi-gas magnetization) as a function of inverse temperature T_F^{**}/T for three coverages of ^{4}He: $D_4 = 2.050$ (open circles), 2.160 (open triangles), and 4.303 (open squares), with T_F^{**} values 126.7, 127.2, and 219.2 mK, respectively. A Curie component is present for $D_4 \leq D_c = 2.66$ layers.

We notice that a sharp peak in T_2 is evident near $D_4 \approx 2.4$ layers, consistent with the presence of a "melting transition" among the ^{3}He; i.e., as the ^{4}He coverage is increased, the ^{3}He become liberated [see Fig. 2(c)]. This may be interpreted as being due to a broad distribution of spin-spin correlation times in the "melted" ^{3}He. This is consistent with a sudden broadening of the excitation spectrum responsible for relaxation. For larger ^{4}He coverages the ^{3}He is fluidlike; for lower coverages some ^{3}He is apparently localized to the solid. This T_2 peak is at lower D_4 for higher temperatures.

For coverages $D_4 > D_c$ a marked increase in the ^{3}He mobility is seen [Fig. 2(a)]. It is expected that at sufficiently low temperature scattering between quasiparticles at the Fermi surface will dominate spin diffusion. Miyake and Mullin [21] have shown that quasiparticle-quasiparticle scattering confined to a two-dimensional Fermi surface exhibits a logarithmic temperature dependence in addition to T^2. In the low-polarization, low-temperature limit, the predicted diffusion coefficient D_{FL} can be expressed in terms of experimental parameters χ, T_F^*, and F_0^a as

$$D_{\mathrm{FL}} = \left(\frac{\chi_0}{\chi}\right)^3 \left(\frac{T_F}{T}\right)^2 \frac{\pi\hbar/m}{|F_0^a|^2 \ln(T_F^*/T)}. \tag{2}$$

To facilitate comparison between this prediction and our diffusion data, we require χ/χ_0 and F_0^a values. In terms of the Fermi-liquid parameters F_1^s and F_0^a,

$$\chi/\chi_0 = m_H \frac{(1 + F_1^s/2)}{(1 + F_0^a)}. \tag{3}$$

To determine the hydrodynamic mass [2] m_H and F_0^a from the χ/χ_0 data, we note that Bhattacharyya, DiPirro, and Gaspirini [1] have measured m^*, $m^* = m_H(1 + F_1^s/2)$, for a ^{3}He coverage of 0.3 monolayer as a function of D_4. Based on our present and earlier [2,5,6] work at five discrete ^{4}He coverages, $D_4 = 3.1$, 3.5, 4.2, 5.2,

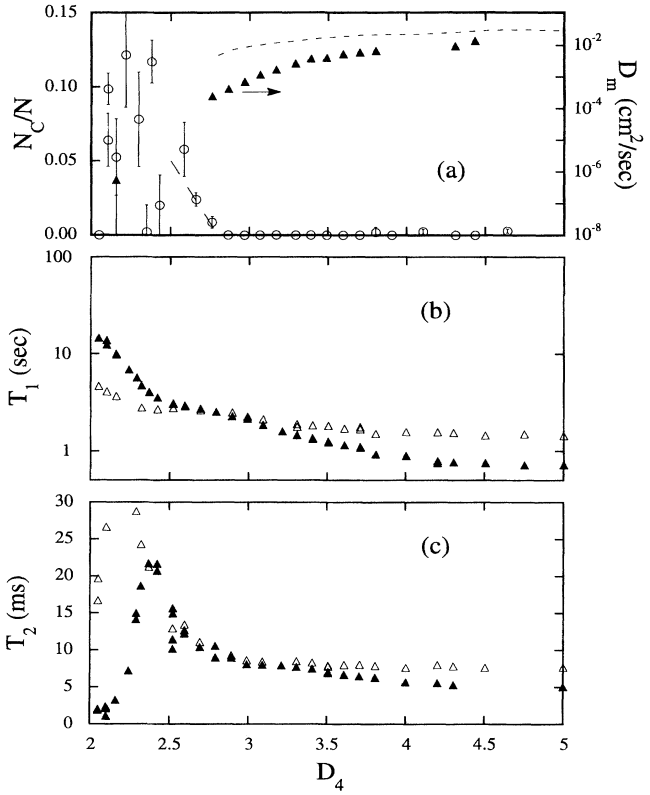

FIG. 2. (a) Diffusion coefficient D_m (triangles) and Curie fraction (circles) as a function of D_4 for $T = 30$ mK. The dashed curve is the Miyake-Mullin [21] prediction using our measurements of χ/χ_0 and deduced F_0^a values. (b) T_1 vs D_4 at 30 mK (closed triangles) and 250 mK (open triangles). (c) T_2 vs D_4 at 30 mK (closed triangles) and 250 mK (open triangles).

and 10.8 layers, the dependence of the magnetization on the ^{3}He coverage is known. For $0.1 \leq D_3 \leq 0.6$, χ/χ_0 is linear in the ^{3}He coverage [2,5] and, in general, extrapolation to zero coverage gives m_H. Since the slopes s of the measured χ/χ_0 vs D_3 vary smoothly over this range of D_4, $1.15 \leq s \leq 1.21$ layer^{-1}, modest interpolation is possible. This interpolation coupled with extensive measurements of χ/χ_0 vs D_4 at $D_3 = 0.1$ layer allows extrapolation of χ/χ_0 to m_H. The ^{3}He density dependence of the Fermi-liquid interactions removed by this process is at most a 2% effect. Figure 3(a) shows the hydrodynamic mass so determined between 2.5 and 7 layers of ^{4}He from two different experimental runs, one with and one without 0.8 layer of O_2 preplated onto the Nuclepore. We point out that this hydrodynamic mass is in good agreement with the calculated trend with coverage which has been predicted [3,4]. Values of F_0^a [see Fig. 3(b)] are obtained as a function of D_4 by combining χ/χ_0, m_H, and m^*. To determine these, we have assumed that F_1^s does not vary greatly with ^{4}He coverage.

The use of χ/χ_0 measured at each ^{4}He coverage and calculated values of F_0^a in the Miyake and Mullin

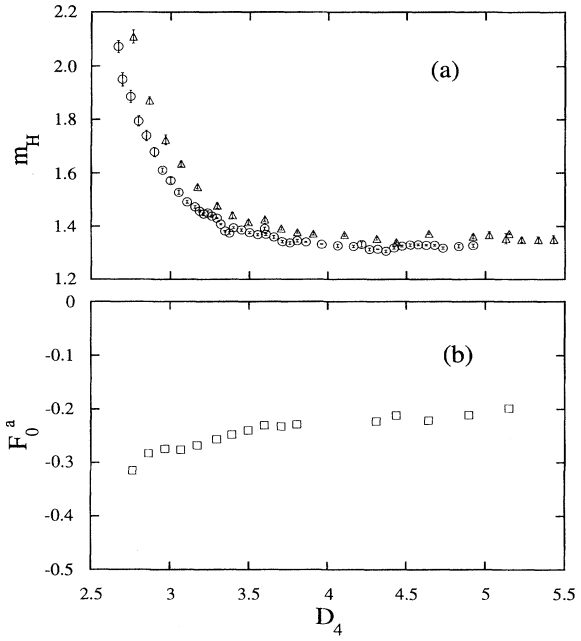

FIG. 3. The hydrodynamic mass (a) and F_0^a (b) as functions of D_4. Different symbols in the case of m_H refer to different runs in which 0.8 monolayer of oxygen was (circles) or was not (triangles) present on the substrate.

[21] expression yields predicted values for the diffusion [Fig. 2(a)]. While the coverage dependence is similar to that of the measured diffusion (primarily due to the role of the hydrodynamic mass in both the susceptibility and the diffusion), the relative magnitudes differ by at least an order of magnitude. Furthermore, the temperature dependence of the measured diffusion is much weaker than $1/T^2$. We find the spin diffusion $D = \alpha D_m$, when scaled by the expected microscopic diffusion D_{FL}, follows a simple power law for temperatures $T \leq 100$ mK [see Fig. 4(a)]:

$$\frac{D(T, D_4)}{D_{\mathrm{FL}}} = \beta T^{\mu}, \tag{4}$$

where for *all* coverages $\mu \simeq 1$. Furthermore the coefficients β can be expressed as $\beta \propto (D_4 - D_c)^{\nu}$, where $\nu \simeq 1$. Thus all of the low-temperature diffusion data follow $D(T, D_4) = \Delta(D_4 - D_c)TD_{\mathrm{FL}}$, where $\Delta = 0.29$ layers^{-1} mK^{-1} [Fig. 4(b)].

This universal scaling may be an indication of percolation. There are three types of disorder to consider. The first is tight binding on the atomic length scale where some fraction of the ^{3}He may be localized and perhaps enter the underlying ^{4}He solid layer. The second is large fluctuations which occur on a much longer length scale and are of sufficient magnitude that in some local regions available states are of higher energy than the ^{3}He chemical potential and so are unoccupied. In this case the surface coverage varies and mobility depends on the topography of the excluded regions and a percolation model is appropriate.

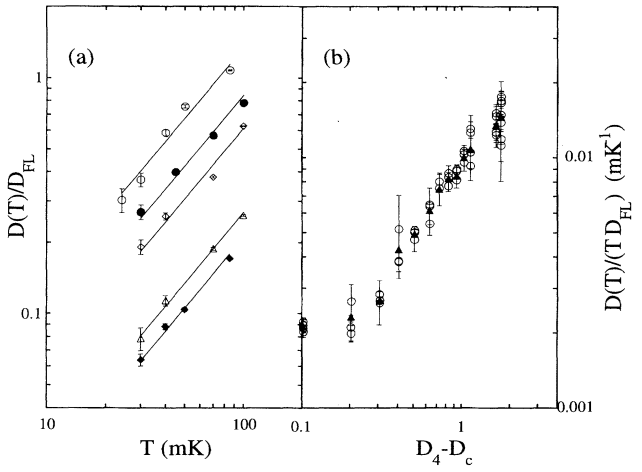

FIG. 4. (a) D/D_{FL} vs T for several coverages: 2.693 (solid diamonds), 2.893 (open triangles), 3.212 (open diamonds), 3.506 (closed circles), and 4.202 (open circles). (b) $D(T)/TD_{\mathrm{FL}}$ vs $D_4 - D_c$ to illustrate the scaling behavior $D(T, D_4) = \Delta(D_4 - D_c)TD_{\mathrm{FL}}$.

Isichenko has noted [11] that when there are fluctuations in which there are no excluded regions but where the two point correlations are comparable to k_F the quasiparticles can be strongly scattered. If at low temperatures the inelastic lifetime is sufficiently long, then weak localization can result [11].

In conclusion, we have reported measurements of the dynamics of the two-dimensional ^{3}He in the environment of a thin ^{4}He film in a context where the fundamental behavior of the substrate can be varied from one of strong to one of weak scattering. A mobility edge is apparent as a function of the ^{4}He coverage. The measured diffusion demonstrates scaling behavior for cases of weak scattering.

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