Properties of 2D ^{3}He on Very Thin ^{4}He Films

P. A. Sheldon* and R. B. Hallock
Laboratory for Low Temperature Physics, Department of Physics and Astronomy, University of Massachusetts at Amherst, Amherst, Massachusetts 01003
(Received 21 June 1996)

We report measurements of the ^{3}He spin diffusion, magnetization, and NMR relaxation times for submonolayer, $n_3 = 0.0064$ Å^{-2} (~0.10 layer), ^{3}He impurities on thin ^{4}He films on Nuclepore. We find a mobility edge, a strong ^{4}He coverage dependence for the ^{3}He ground state energy, and the absence of an excited state for the ^{3}He for very low ^{4}He coverages. A $\sim 10^3 - 10^4$ increase in the value of the diffusion coefficient occurs over a narrow ^{4}He coverage range, $0.15 \leq n_4 \leq 0.23$ Å^{-2}, and a large Curie-like component is present in the magnetization for $n_4 \leq 0.20$ Å^{-2}. [S0031-9007(96)01347-6]

PACS numbers: 67.70.+n, 67.60.–g, 76.60.–k

At low temperature, submonolayer ^{3}He atoms on a superfluid ^{4}He film occupy a surface state [1] at the free surface of the film and behave as a nearly ideal two-dimensional Fermi gas, exhibiting degenerate magnetization [2]. In the dimension perpendicular to the substrate, the ^{3}He constitute a system akin to particles in a box, and the energy of the ground state and the first excited state has been measured [3–5] as a function of ^{4}He coverage and found to be in reasonable agreement with theoretical predictions [6,7]. For such a system, the ^{3}He are relatively free to move along the superfluid ^{4}He surface. On strong-binding surfaces, approximately two layers of solid ^{4}He are found beneath the superfluid, adjacent to the substrate. In this Letter we use NMR techniques to explore the behavior of the ^{3}He in the ^{4}He coverage regime near and below that necessary for superfluid behavior. As the ^{4}He coverage is reduced, the potential which holds the surface state is expected to change shape, altering the energetics of the ^{3}He. For such a thin film, random disorder in the potential experienced by the ^{3}He atoms is introduced by the surface roughness of the substrate and the semisolid ^{4}He film adjacent to it. The strength of this disorder is tunable by variation of the ^{4}He film coverage. If the disorder is strong enough, localization is expected to occur due to coherent backscattering from the random potential. This type of localization was first recognized by Anderson [8] in electronic systems, and it is expected to be present for any wave phenomena [9]. For a Fermi system, as $T \to 0$ the localization of the fermion wave function is predicted to result in a diverging Curie-like component to the magnetization due to the localized fermions no longer being part of the degenerate liquid. This localization, and consequent hindered mobility, should give rise to a greatly reduced diffusion coefficient, D [10]. In the experiments we report here, NMR measurements are used to determine D, the magnetization, M, and the relaxation times, T_1 and T_2, for the ^{3}He in the thin ^{4}He film environment, as a function of temperature and ^{4}He coverage [11], D_4. Each of these quantities is found to demonstrate behavior which, at low ^{4}He coverages, is a strong function of the ^{4}He film coverage, consistent with localization. We also find that for ^{4}He coverages below the minimum coverage necessary for superfluidity, the ground state energy increases as the ^{4}He coverage is reduced, and the excited state for the ^{3}He disappears.

Sprague *et al.* [4,12,13] reported results of NMR measurements for 0.1 layer ^{3}He on a thin ^{4}He film which explored the properties of the ^{3}He in the film and which give some support to the physical picture of localization described above. Although the data were limited, for a ^{4}He film of coverage $D_4 \leq 2.7$ (bulk-density) layers their magnetization measurements show evidence for nondegenerate behavior at the lowest temperatures studied, indicating that the magnetization may contain a Curie component from localized spins. Their measurements also showed some evidence that the spin-diffusion coefficient increases strongly over a narrow range of ^{4}He film coverage. Measurements of the ^{4}He coverage dependence of the relaxation time T_2 showed a maximum at low coverage, which was interpreted as behavior consistent with a melting transition.

In the work we report here [14], which provides a much more thorough exploration of the low ^{4}He coverage regime, we used pulsed NMR techniques at 62.9 MHz to measure D, M, T_1, and T_2 for thin mixture films with ^{4}He coverages $1.89 \leq D_4 \leq 2.90$ bulk-density layers ($0.15 \leq n_4 \leq 0.23$ Å^{-2}), with a fixed ^{3}He coverage of $D_3 = 0.10$ layer ($n_3 = 0.0064$ Å^{-2}) and for temperatures $40 \leq T \leq 500$ mK. The relatively strong-binding substrate which supports the ^{4}He is Nuclepore, a polycarbonate material threaded by $\sim 3 \times 10^8$, 200 nm diam pores/cm^2, which provides surface area for NMR signals of reasonable signal to noise levels. The helium films we study here are thin enough to ensure the absence of capillary condensation [15] in the Nuclepore. The ^{4}He surface underlying the ^{3}He ranges from solid to fluid to superfluid over this ^{4}He coverage range. A third sound resonator is present in the cell along with the NMR resonator, and this is used to confirm coverage changes where appropriate when helium is added to the cell, and to determine the ^{4}He coverage at which the superfluid transition occurs. Magnetization and relaxation times are measured with Hahn spin echoes, and longitudinal spin diffusion is measured in a static field gradient with stimulated echoes. During the evolution of this experiment, we began at the lowest ^{4}He coverage, and carried out our measurements at selected temperatures following incremental additions of ^{4}He for a fixed amount of ^{3}He. This protocol was necessary since the removal of helium from the sample chamber causes the concentration to change in an unknown manner.

For $T > 300$ mK, evaporation of ^{3}He from the NMR coil occurs, and this provides for a determination of the ground state energy, ϵ_0, for the ^{3}He using the method described earlier by Sprague *et al.* [4]. Studies of the temperature dependence of T_1, which has been shown [4,5] to be of the form $1/T_1 = A + B/T^{1/2} + C\exp(-\Delta/T)$, are used to measure the energy gap, Δ, between the Fermi energy, ϵ_F, and the first excited state, ϵ_1, for the ^{3}He in the film. Our measurements of M and T_1 in the low coverage regime yield a determination of ϵ_0, Δ, and ϵ_F, and thus we deduce ϵ_1. We find an absence of thermally activated behavior for T_1 for $D_4 < 2.49$ layers $= D_A$, which implies that the excited state ceases to exist in this coverage range. Our results along with earlier data obtained at higher coverages are shown in Fig. 1. ϵ_0 and ϵ_1 increase with decreasing coverage, with ϵ_1 apparently reaching zero for $D_4 \approx D_A$. This suggests that the surface state potential becomes more narrow as the film thins. Also shown on Fig. 1 are the results of theoretical calculations for the lowest energy states of the ^{3}He in the film, with the coverage scales shifted so as to correspond to that of the experiment. In the density functional calculation [6], the theory assumes two solid layers (with coverage 0.108 and 0.078 Å^{-2}) which has been accounted for in affixing the theory to our coverage axis. In the microscopic calculation [7], which provides an upper limit to the energy values, the

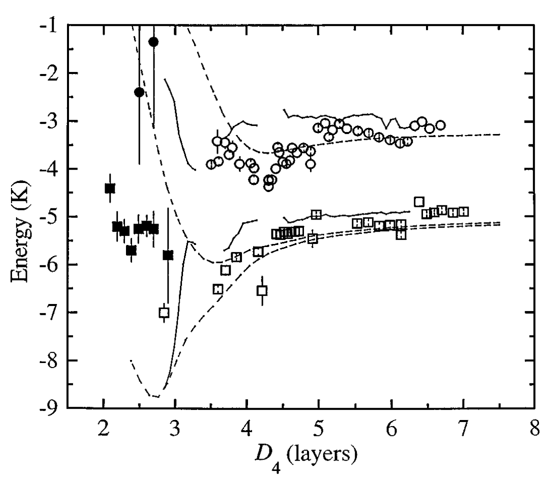

FIG. 1. Ground state (squares) and first excited state (circles) energy values determined in this work (solid symbols) and by Sprague *et al.* [4] (open symbols). No evidence for the existence of the excited state is found for $D_4 < 2.49$ bulk-density layers. The dashed curve is a density functional prediction due to Treiner [6], and the solid curve is a microscopic prediction due to Krotscheck [7].

calculation starts at the interface between the solid and the fluid, and we have added $D_i = 2.41$ bulk-density layers to this calculation to affix the theory to our axis. This value of D_i for the coverage of the solidlike layer comes from an examination of the ^{4}He coverage dependence of the magnetization to be described shortly. It is also consistent with an examination of the coverage dependence of the relaxation time T_1 made in a manner reminiscent of the technique used by Swanson *et al.* [16] to identify monolayer completion. This D_i is a bit lower than the value 2.66 used by Sprague *et al.* [12], presumably due to the presence of a somewhat different protocol used to create the sample. There is general agreement with the theory for the coverage dependence of ϵ_0 and ϵ_1, with the predictions consistent with the data on the interpretation that the first excited state disappears at low coverages.

The longitudinal spin diffusion D is measured with stimulated pulse-echo sequences, which allows one to observe diffusion over a time scale of $T_1 \simeq 200T_2$. The stimulated echo sequence is $\pi/2$-τ_2-$\pi/2$-τ_1-$\pi/2$ echo, where $\tau_2 \sim T_2$ and $\tau_1 \sim T_1$. To measure D we utilize the time τ_1 and the magnetic field gradient, G, dependence of the stimulated echo height, $E(\tau_1, G)$, where

$$E(\tau_1, G) = \frac{M_0}{2}\exp\left[-\frac{\tau_1}{T_1} - \frac{\gamma^2 G^2 D \tau_2^3}{3}\left(\frac{3\tau_1}{\tau_2} + 1\right)\right], \tag{1}$$

and where γ is the gyromagnetic ratio. The stimulated echo is measured for at least four different values of τ_1 and G, and the slopes of the τ_1 dependence of $\ln(E)$ for each G are fit linearly by G^2. The resulting slope allows a determination of D. These stimulated echo measurements probe time scales long enough for the ^{3}He spins to move among pores, hence the measured spin diffusion D is expected to be related to the bare spin diffusion D_{bare} through the tortuosity factor for Nuclepore, $\alpha = 16$ [17], so that $D_{\text{bare}} = \alpha D$. The ^{4}He coverage dependence of D is shown for four temperatures along with earlier data [12,18] in Fig. 2. Between 2 and 3 layers of ^{4}He, D increases smoothly by 10^3–10^4. The dramatic increase in mobility over such a small coverage range is reminiscent of the mobility edge for electrons seen in thin metal films [19]. The temperature dependence of D gets weaker with increasing coverage. In Fig. 2 (inset) power law fits, $D = AT^{\beta}$, which yield $\beta = 0.7$ and 0.5 for $D_4 = 2.19$ and 2.39 layers, respectively, are shown with the data. At $D_4 = 2.91$, D shows very weak temperature dependence. This behavior is in contrast to the temperature dependence seen at higher ^{4}He coverages, $D_4 > 3$ layers, where Sprague *et al.* [12,18] found that power law fits for $T \le 150$ mK resulted in exponents $-1 \lesssim \beta \lesssim -1.5$. Localization of the ^{3}He should result in a disappearing diffusion constant at low temperature. For our lowest ^{4}He coverages, we find D decreases with decreasing T, a result which is consistent with the expectation that $D \rightarrow 0$ if the ^{3}He is completely localized.

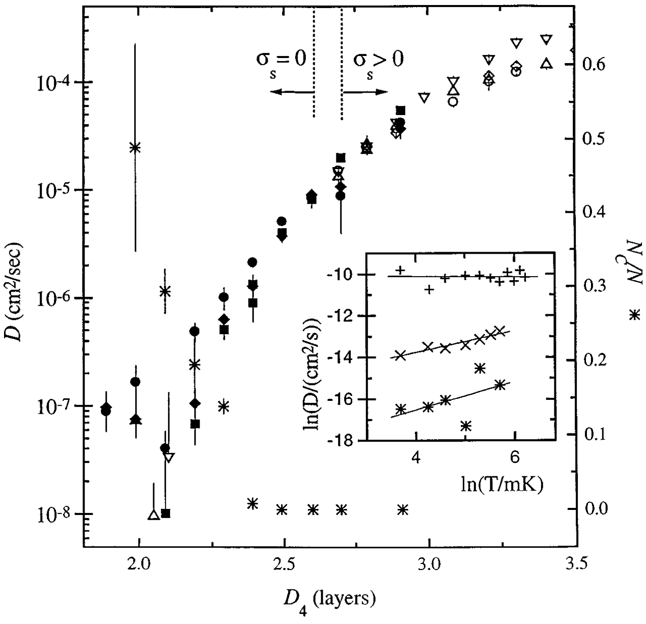

FIG. 2. D as a function of ^{4}He coverage for various temperatures for this work (solid symbols), and from Ref. [3] (open symbols). Inverted triangles, 30 mK; squares, 40 mK; diamonds, 100 mK; triangles, 150 mK; and circles, 200 mK. Also shown are N_C/N values (asterisks) for this work. The superfluid transition occurs in the range $2.6 < D_4 \leq 2.7$ layers. The inset shows power law fits (solid lines) to the temperature dependence of the diffusion constant for three representative coverages, 2.19 (asterisk), 2.39 (cross), and 2.91 (plus) bulk-density layers.

Also noted in Fig. 2 is the coverage range, $D_4 \geq 2.70$ layers where the superfluid density is nonzero as determined by the third sound measurements. Third sound was searched for at $T = 100$ mK for each coverage, $D_4 \geq 2.49$ layers. It was first observed at $D_4 = 2.70$ layers, but not seen at $D_4 = 2.60$ layers, which indicates that at $T = 100$ mK the onset of superfluidity occurred at D_c, where $2.60 < D_c \leq 2.70$ layers. Kosterlitz–Thouless theory [20] (KT theory) predicts that the ratio of the areal superfluid density to the temperature is a universal constant at the superfluid transition, $(\sigma_s/T)_c = (2k_B/\pi)(m_4/\hbar)^2$. For $T \leq 500$ mK, $\rho_s/\rho \simeq 1$. The KT theory predicts the transition at a ^{4}He coverage given by $(d_s/T)_c = 0.68$ layer/K, where d_s is the superfluid coverage measured above the inert ^{4}He layer closest to the substrate. We measured the inert layer to be of coverage $D_i = 2.41 \pm 0.09$ layers, where Sprague *et al.* [12,18] found $D_i = 2.66 \pm 0.03$ layers. Thus, at $T = 100$ mK, for $D_i = 2.41$ layers, we would predict that the superfluid transition will occur at a ^{4}He coverage of $D_c^* = 2.48 \pm 0.11$ layers, which is reasonably consistent with our observations. As shown in Fig. 2, the superfluid transition has no noticeable effect on the ^{3}He spin-diffusion coefficient. For $D_4 \leq D_c$, the ^{4}He is a combination of solid and liquid, but not superfluid. As the coverage is reduced below D_c the ^{3}He atoms apparently become increasingly exposed to irregularities imposed by the solidlike ^{4}He and by the substrate, and D decreases.

Magnetization vs inverse temperature is shown in Fig. 3 for $D_3 = 0.1$ layer for the ten ^{4}He coverage values studied. At the lowest temperatures and highest ^{4}He coverages the magnetization is degenerate. A large Curie component to the magnetization is present for the lowest ^{4}He coverages. The magnetization data can be represented by the expression for Pauli paramagnetism, augmented by an additional term, C/T, which represents [12] a ^{4}He coverage-dependent Curie contribution,

$$M = \frac{C_0}{T_F^{**}}\left[1 - \exp\left(-\frac{T_F^{**}}{T}\right)\right] + \frac{C}{T}, \tag{2}$$

where C_0 is the Curie constant and T_F^{**} is the degeneracy temperature. $N_C/N = C/(C + C_0)$ is the Curie fraction, the fraction of spins which contribute to the Curie component of the magnetization. The Curie fraction determined from fits of the data by Eq. (1) is shown vs ^{4}He coverage in Fig. 2. The Curie fraction is large at low coverage and decreases with increasing coverage, with $N_C/N = 0$ for $D_4 \geq 2.49$ layers. This behavior is consistent with the ansatz that for low ^{4}He coverages a fraction of the ^{3}He atoms is constrained due to the roughness provided by the solidlike ^{4}He, and thus these atoms are localized [12].

We confirm the behavior previously seen for the transverse and longitudinal relaxation times [5,12,18]. For all temperatures studied in the range $40 < T < 200$ mK, a maximum is observed in T_2 for $D_4 \leq 2.3$ layers. The maximum gets larger and moves to lower coverages as the temperature is increased. For $D_4 < 2.4$ layers, T_1 rises with decreasing film coverage and shows strong temperature dependence at low coverages. For $D_4 \geq 2.4$ layers,

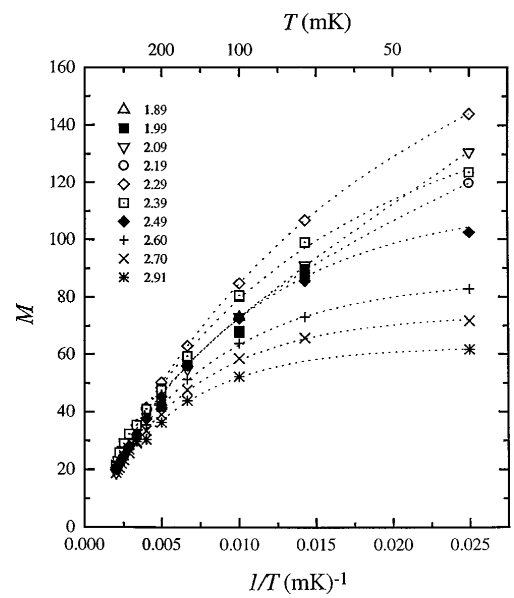

FIG. 3. Magnetization (arbitrary units) versus inverse temperature for various D_4 (bulk-density layers) with fixed ^{3}He coverage, $D_3 = 0.1$ layer. Dashed lines are fits to the Pauli paramagnetism plus a Curie component, Eq. (2).

much weaker temperature dependence for T_1 is observed. Since a new solid ^{4}He surface was created for this experiment, detailed differences in the dynamics observed at low ^{4}He coverage between our data and the earlier work [12] are likely due to the differences in roughness and randomness of the ^{4}He solid surface. At higher coverages, Alikacem *et al.* [3] found that the temperature dependence of T_1 is independent of ^{4}He coverage, thus they concluded that T_1 is dominated by processes at the film surface. At lower coverages, the temperature dependence of T_1 has a strong dependence on coverage, implying that processes such as relaxation with paramagnetic impurities in the substrate may dominate when the ^{3}He gets close enough to the substrate. If relaxation is dominated by interactions with the substrate, then it is expected [21] that the relaxation rate will depend on the diffusion coefficient as $1/T_1 \propto D$. This is not observed. We observe a less than 1 order of magnitude increase in T_1 for $2.4 \geq D_4 \geq 1.89$ layers. This behavior does not scale as $1/T_1 \propto D$ since we observe a 2 orders of magnitude decrease for D over the same coverage range. Since the proportionality should be dependent on geometry, and we expect the effective geometry of the film surface to be changing over this coverage range, this behavior is perhaps understandable.

In summary, the magnetization, relaxation times, and diffusion have been measured for a ^{3}He impurity on a low coverage ^{4}He film. The Curie fraction goes to zero for $D_4 \sim 2.4$ layers, near the ^{4}He coverage at which the peak is observed in T_2. The decrease in T_2 and the vanishing Curie fraction are consistent with a melting transition in the ^{3}He [12]. We observe a large decrease in mobility with decreasing ^{4}He coverage, and the temperature dependence of the diffusion constant is consistent with $D \rightarrow 0$ at low temperatures. In addition, the ground state energy is observed to become increasingly less negative as the ^{4}He coverage is reduced, and the excited state disappears, which suggests that the potential available to the ^{3}He becomes more narrow as the ^{4}He coverage is reduced. We conclude that the structure observed in the magnetization, relaxation times, and diffusion constant is consistent with the localization of a fraction of the ^{3}He by the inhomogeneities of the substrate and immobile ^{4}He surface for $D_4 \leq 2.39$ layers. It is not clear to us whether this should be interpreted as localization of the coherent backscattering type (with ^{3}He localized in a surface state at the free surface of the film) or as localization with ^{3}He trapped as a part of the solid layer. Given the evolution of the binding energy with coverage, the former seems more likely.

We thank E. Krotscheck and J. Treiner for the use of unpublished theoretical predictions for ϵ_0 and ϵ_1, and D.T. Sprague for helpful comments. This work was supported by the NSF through DMR 91-22348 and DMR 94-22208.

*Present address: Davidson College, Davidson, NC.

[1] F.M. Gasparini, B. Bhattacharyya, and M.J. DiPirro, Phys. Rev. B **29**, 4921 (1984); A.F. Andreev, Zh. Eksp. Teor. Fiz. **50**, 1415 (1966) [Sov. Phys. JETP **23**, 939 (1966)]; D.S. Sherrill and D.O. Edwards, Phys. Rev. B **31**, 1338 (1985).
[2] J.M. Valles, R.H. Higley, R.B. Johnson, and R.B. Hallock, Phys. Rev. Lett. **60**, 428 (1988).
[3] N. Alikacem, D.T. Sprague, and R.B. Hallock, Phys. Rev. Lett. **67**, 2501 (1991).
[4] D.T. Sprague, N. Alikacem, P.A. Sheldon, and R.B. Hallock, Phys. Rev. Lett. **72**, 384 (1994).
[5] R.B. Hallock, in *Progress in Low Temperature Physics*, edited by W.P. Halperin (Elsevier Science BV, Amsterdam, 1995), Vol. XIV, p. 321.
[6] N. Pavloff and J. Treiner, J. Low Temp. Phys. **83**, 331 (1991); (private communication).
[7] E. Krotscheck (to be published); (private communication).
[8] P.W. Anderson, Phys. Rev. **109**, 1490 (1958).
[9] P.W. Anderson, Philos. Mag. B **52**, 505 (1985); see, for example, S. He and J.D. Maynard, Phys. Rev. Lett. **57**, 3171 (1986); D.T. Smith, C.P. Lorenson, R.B. Hallock, K.R. McCall, and R.A. Guyer, Phys. Rev. Lett. **61**, 1286 (1988); D.T. Smith, C.P. Lorenson, and R.B. Hallock, Phys. Rev. B **40**, 6648 (1989).
[10] R.N. Bhatt and D.S. Fisher, Phys. Rev. Lett. **68**, 3072 (1992); P.A. Lee and T.V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985); D. Belitz and T.R. Kirkpatrick, Phys. Rev. B **44**, 955 (1991).
[11] Here we adopt a coverage scale measured in bulk-density-equivalent atomic layers, D_4. With this convention, $D_4 = N_4/23.0 = 12.8n_4$, where N_4 is the ^{4}He coverage in micromoles, and n_4 is the coverage in atoms/Å^2, with $n_4 = 0.00340N_4$.
[12] D.T. Sprague, N. Alikacem, and R.B. Hallock, Phys. Rev. Lett. **74**, 4479 (1995).
[13] D.T. Sprague, N. Alikacem, P.A. Sheldon, and R.B. Hallock, Physica (Amsterdam) **194-196B**, 631 (1994).
[14] A preliminary report has appeared: P.A. Sheldon and R.B. Hallock, in Proceedings of the 21st International Conference on Low Temperature Physics [Czech. J. Phys. **46**, S1, 425 (1996)].
[15] J.M. Valles, Jr., D.T. Smith, and R.B. Hallock, Phys. Rev. Lett. **54**, 1528 (1985).
[16] D.R. Swanson, D. Candela, and D.O. Edwards, J. Low Temp. Phys. **72**, 213 (1988).
[17] S.M. Cohen, R.A. Guyer, and J. Machta, Phys. Rev. B **33**, 4664 (1986).
[18] D.T. Sprague, Ph.D. dissertation, University of Massachusetts at Amherst, 1993 (unpublished).
[19] N. Mott, M. Pepper, S. Pollitt, R.H. Wallis, and C.J. Adkins, Proc. R. Soc. London A **345**, 169 (1975); J.M. Valles, Jr., A.E. White, K.T. Short, R.C. Dynes, J.P. Garno, F.F.J. Levi, M. Anzlowar, and K. Baldwin, Phys. Rev. B **39**, 11 599 (1989).
[20] J.M. Kosterlitz and D.J. Thouless, J. Phys. C **6**, 1181 (1973); D.R. Nelson and J.M. Kosterlitz, Phys. Rev. Lett. **39**, 1201 (1977).
[21] M. Lowe, P.C. Hammel, R.E. Ecke, K. Bedell, and M. Takigawa, Phys. Rev. B **37**, 2281 (1988).