Energetics of ^{3}He States in ^{4}He Films

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We report measurements of the NMR spin-lattice relaxation time, T_1, of ^{3}He in ^{4}He films for $0.03 < T < 0.60$ K as a function of ^{4}He coverage of 0.1 layer of adsorbed ^{3}He. The results show thermally activated behavior for $T > 0.25$ K and yield the energy difference, $\delta\epsilon$, between the ^{3}He ground state and the first excited state; $\delta\epsilon$ shows substantial structure as a function of ^{4}He coverage.

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^{3}He atoms bound to a thin ^{4}He film constitute a system of remarkable richness. As in the case for bulk ^{4}He [1], the lowest state available to a single ^{3}He atom in a ^{4}He film is one in which the ^{3}He resides at the surface of the ^{4}He film [2,3]. This is due to the combined effect of the substrate potential and ^{3}He-^{4}He interactions. In the dilute limit the ^{3}He atoms in the surface state form a quasi-two-dimensional gas [4–7] which is degenerate for temperatures below the Fermi temperature. The effective potential defined by the substrate and the ^{4}He also produce bound states of higher energy for the ^{3}He in the film [4–7]. This spectrum of states is expected to be sensitive to the ^{4}He coverage, n_4 [3]. For the present work, we have used NMR to study the dynamics of the ^{3}He in mixture films and by this means have measured the energy separation, $\delta\epsilon$, between the ground and first excited state of the ^{3}He as a function of n_4. We find that $\delta\epsilon$ shows substantial structure.

In these experiments, we have carried out measurements of the spin-lattice relaxation time T_1 of ^{3}He on a superfluid ^{4}He film as a function of ^{4}He coverage n_4 for $0.27 < n_4 < 0.53$ atom/Å^2 [8], and temperature T for $0.03 < T < 0.60$ K. The ^{3}He coverage is fixed at $n_3 = 0.0066$ atom/Å^2 ($\sim$0.1 layer). Such a ^{3}He system is well described as a 2D weakly interacting Fermi liquid [4–7] with Fermi temperature $T_F \sim 0.24$ K. In earlier work for $T < 0.25$ K, the measured relaxation rate $T_1^{-1}(T)$ was observed [8] to behave as $T_1^{-1}(T) \sim A + B/\sqrt{T}$. Here, for $T > 0.25$ K, we find $T_1^{-1} \sim \exp(-\Delta/T)$ with characteristic energy Δ. We interpret Δ to be the energy gap between the Fermi level of the ^{3}He in the ground state and an excited state of the ^{3}He in the film [4].

The substrate used for this experiment is Nuclepore [9], polycarbonate filter material, which provides most of the 1.77 m^2 ($\pm$ 10%) sample cell surface area. The measurements we report here are made by use of pulsed NMR [6] in a 2-T external field ($\omega/2\pi \sim 62.9$ MHz). The temperatures are achieved by use of a dilution refrigerator and measured with a Speer 100-Ω carbon resistor previously calibrated against a ^{3}He melting curve thermometer [10]. The spin-lattice relaxation time T_1 is measured by spin-echo techniques using a series of $\frac{1}{2}\pi$-τ-$\frac{1}{2}\pi$-τ_0-π rf-pulse sequences as a function of τ for $\tau_0 = 0.3$ msec. The amplitude of the spin echo from the ^{3}He system is directly proportional to the magnetization which has recovered after a time τ following the first $\frac{1}{2}\pi$ rf pulse. For the range of ^{4}He coverages and temperatures discussed here, the decay of the spin-echo height with τ is well described by a single exponential over one to two decades in echo height with a characteristic time T_1. The magnetization and the spin-spin relaxation time T_2 are measured by use of $\frac{1}{2}\pi$-τ-π spin-echo pulse sequences.

Figure 1 shows measurements of the spin-lattice relaxation rate T_1^{-1} as a function of $1/\sqrt{T}$ ($0.03 < T < 0.60$ K) for several ^{4}He coverages. Two temperature regions can be identified. For $T < 0.25$ K, $T_1^{-1}(T)$ is a linear function [8] of $1/\sqrt{T}$; $T_1^{-1} = A + B/\sqrt{T} \equiv W_{LT}$. The coefficient B has been shown to have intriguing n_4 dependence which apparently is correlated with the amount of

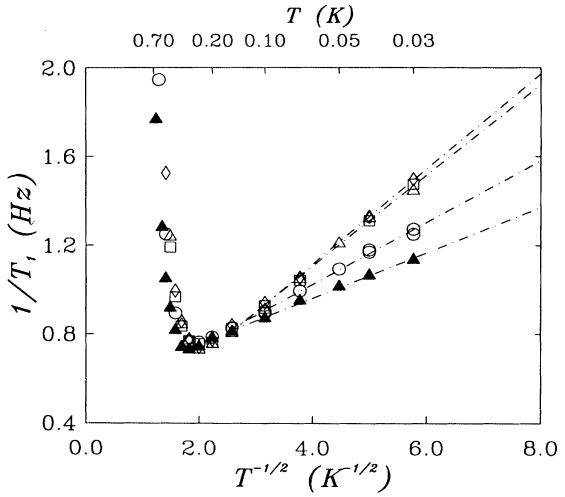

FIG. 1. T_1^{-1} vs $T^{-1/2}$ for various n_4. For $T < 0.25$ K, $T_1^{-1}(T) \sim A + B/\sqrt{T}$. The dot-dashed lines are straight-line fits for $T_1^{-1} \sim T^{-1/2}$. For $T > 0.25$ K, $T_1^{-1}(T)$ increases exponentially with temperature. Coverages: 0.217 Å^{-2} (solid triangles), 0.290 Å^{-2} (circles), 0.339 Å^{-2} (diamonds), 0.362 Å^{-2} (squares), and 0.400 Å^{-2} (open triangles).

superfluid in the ^{4}He film [8]. For $T > 0.25$ K, $T_1^{-1}(T)$ increases dramatically with temperature. For simplicity, we assume that the observed deviation of the relaxation rate from W_{LT} is due to the addition of another relaxation rate, W_{HT}, which is associated with the mechanisms of relaxation for $T > 0.25$ K. Thus $T_1^{-1} \sim W_{LT} + W_{HT}$.

We find that the rate W_{HT} is well described by an exponential, $W_{HT} \sim \exp(-\Delta/T)$, as shown in Fig. 2, with Δ dependent on n_4. The values of W_{HT} shown in Fig. 2 are obtained from data like that shown in Fig. 1 by subtraction: $W_{HT} = T_1^{-1}(T) - W_{LT}$, where we assume W_{LT} retains its $\sqrt{T}$ character even for $T > T_F$ [11]. This exponential behavior of W_{HT} should not be confused with the exponential behavior seen by others [12,13] in $T_1(T)$ in the *gas* phase, which is dominated by wall relaxation. Here, W_{HT} is not due to the evaporation of ^{3}He from the mixture film. Measurements of the magnetization $M(T)$ as a function of temperature show that the evaporation process is observable for $T \geq 0.60$ K with a measured [14] binding energy $\sim 6 \pm 1$ K, which is substantially larger than the average value of $\Delta \sim 1.8$ K (our result for the binding energy is consistent with the known value ~ 5 K [13,14]). Furthermore, since T_1 of the vapor phase of ^{3}He is longer than that of the film, the evaporation process is expected to cause the relaxation rate of the film to *decrease* exponentially [13,14] with increasing temperature; this is contrary to the behavior observed in Fig. 2.

To enhance our understanding of this relaxation rate $T_1^{-1}(T)$, we assume that $W_{LT} \sim n_0 W_0$, where n_0 and W_0 are respectively the temperature-dependent density and relaxation rate of the ^{3}He spins in the ground state; $W_0(T)$ retains the form [8] $a + b/\sqrt{T}$. This is a reasonable assumption since, generally, the bulk relaxation rate T_1^{-1} is proportional to the density of ^{3}He for classical or degenerate ^{3}He liquid [15]; this proportionality is consistent with earlier measurements of T_1 done in mixture films for ^{3}He coverages $0.1 < d_3 < 0.5$ layer [7]. Similarly, we take $W_{HT} \sim n_1 W_1$, where n_1 and W_1 are respectively the density and the relaxation rate of ^{3}He spins in the excited state. As the temperature is increased, a fraction of the ^{3}He spins are thermally promoted into an excited state in the film, providing an additional channel for relaxation (Fig. 3). Assuming that the exchange rate W_E between the two states is faster than W_0 and W_1 [16], $T_1^{-1}(T) \sim n_0 W_0 + n_1 W_1$.

Next, we investigate the temperature dependence of n_0 and n_1. By modeling the film to have discrete energy levels ϵ_0 and ϵ_1, we can solve [17] for the chemical potential of the ^{3}He, using a Fermi distribution, at fixed number N, and energy separation $\delta\epsilon = \epsilon_1 - \epsilon_0$. This yields n_0 and n_1 as a function of temperature. N is the total number of ^{3}He atoms, $N/A = n_0 + n_1$; A is the surface area. In the temperature range of the measurements done here, $n_0(T) \sim 1 - \exp(-\Delta/T)$ whereas $n_1(T) \sim \exp(-\Delta/T)$; Δ is approximately the energy separation between the Fermi level and the excited state. Consequently, the observed relaxation rate can be written as $T_1^{-1}(T) \simeq W_0(T) + W_1(T)\exp(-\Delta/T)$ [18]. The rate W_1, which characterizes the mechanisms of relaxation from the excited state, may be a function of temperature. However, any anticipated temperature dependence in W_1 is weak compared with the exponential behavior of $n_1(T)$. Thus, for the purpose of extracting the energies Δ, we assumed that $W_1(T)$ is independent of temperature [19]. We find that the relaxation rate in the excited state is typically 50 times larger than that of the ground state, $W_1 \sim 50 W_0$ [20].

Within the context of this model, the slopes of Fig. 2 provide a direct measure [21] of the energy separation Δ

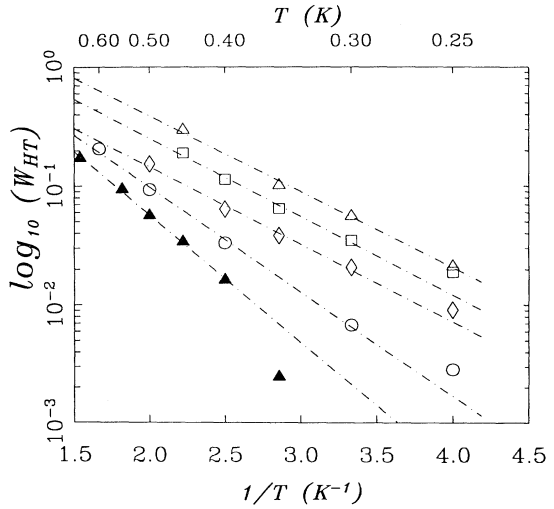

FIG. 2. $\log_{10}(W_{HT})$ vs $1/T$ for several ^{4}He coverages. $W_{HT} \sim \exp(-\Delta/T)$. Δ is given by the slopes of the straight-line fits (dot-dashed lines) for $\log_{10}(W_{HT}) \sim 1/T$. Coverages: 0.217 Å^{-2} (solid triangles), 0.290 Å^{-2} (circles), 0.339 Å^{-2} (diamonds), 0.362 Å^{-2} (squares), and 0.400 Å^{-2} (open triangles).

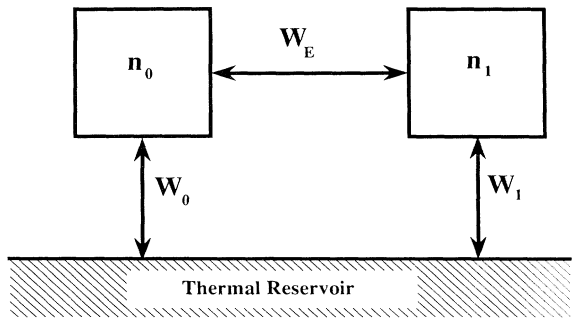

FIG. 3. Schematic illustration of the two states available to the ^{3}He. n_0 and W_0 are respectively the density and the relaxation rate of the ^{3}He spins in the ground state. n_1 and W_1 are respectively the density and the relaxation rate of the ^{3}He spins in the first excited state. The exchange rate between the two states, W_E, is assumed to be much greater than either W_0 or W_1.

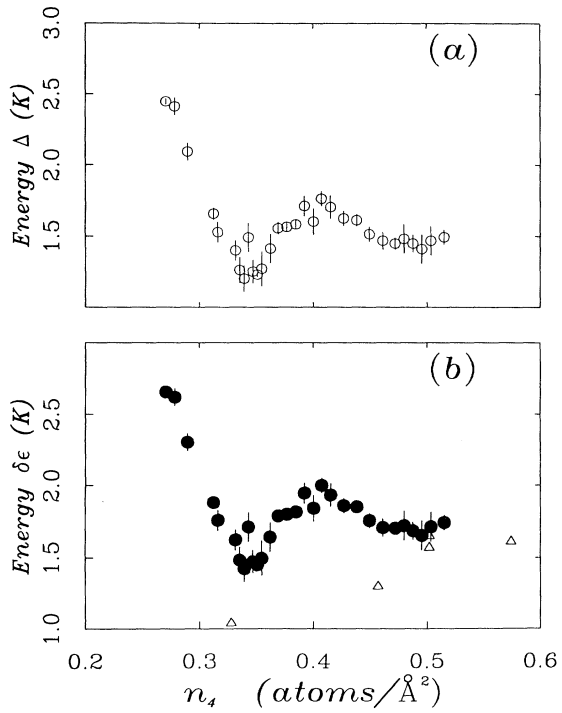

FIG. 4. (a) Δ vs n_4. Δ is the energy separation between the Fermi level of the ^{3}He in the ground state and the first excited state. The evolution of $\Delta(n_4)$ shows a minimum of 1.22 K at $n_4 \simeq 0.34$ Å^{-2}. (b) $\delta\epsilon$ vs n_4. $\delta\epsilon$ is the energy separation between the ground and the first excited state of the ^{3}He in the film. The solid circles are the energies obtained using NMR techniques. $\delta\epsilon(n_4)$ shows a minimum of -1.5 K; at our highest ^{4}He coverage $\delta\epsilon \sim 1.7$ K. The ^{3}He coverage ~ 0.1 layer. Also shown here are energies as given from heat-capacity measurements (triangles) for $d_3 \simeq 0.3$ layer from Ref. [4].

between the Fermi level and the excited state in the film. In Fig. 4(a), we present the energies Δ as a function of ^{4}He coverage n_4. Δ has considerable structure as a function of n_4; Δ has a minimum for $n_4 \simeq 0.34$ atom/Å^2 and a local maximum for $n_4 \simeq 0.41$ atom/Å^2. By adding the Fermi energy to Δ, we obtain the energy separation between the ground and the excited state $\delta\epsilon \equiv \epsilon_1 - \epsilon_0$. To accomplish this, we note that the magnetization is found to be well described by a 2D ideal Fermi gas, with a degeneracy temperature which shows a weak ^{4}He-coverage dependence. This degeneracy temperature is within 10% of the Fermi temperature T_F [4]. Consequently, we approximate the Fermi temperature at each ^{4}He coverage by the degeneracy temperature. Figure 4(b) shows the variation of this energy separation, $\delta\epsilon$, as a function of ^{4}He coverage.

The values for the excitation energy $\delta\epsilon(n_4)$ have been obtained on a much finer n_4 grid than the earlier heat-capacity results of Bhattacharyya, DiPirro, and Gasparini [4] [Fig. 4(b)] which were at a somewhat higher ^{3}He coverage, $d_3 \simeq 0.3$ layer. When one accounts for the difference in ^{3}He coverage, the absolute values of the energies are quite consistent for the two different techniques.

These data should be useful in testing emerging theories for ^{3}He in mixture films. Recent theoretical work by Pavloff and Treiner [3] and by Epstein and Krotscheck [22] has explored the properties of ^{3}He in ^{4}He films. In the case of thin ^{4}He films, these workers predict structure in the evolution of the energy levels ϵ_i with n_4; this is generally consistent with the energetics we report here. Each calculation yields a local minimum in $\delta\epsilon$ qualitatively similar to our observations. In the limit of large n_4, Pavloff and Treiner predict two bound states at the ^{4}He surface with $\delta\epsilon \simeq 1.8$ K; the first excited state is expected to remain at the film surface for modest ^{3}He coverage. In the same limit Epstein and Krotscheck predict a single surface state for which the energy difference between this surface state and the lowest state in the ^{4}He film is $\simeq 1.4$ K. The agreement between the theory and our measurements is encouraging. Finite ^{3}He coverage calculations are needed.

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[1] D. O. Edwards and W. F. Saam, in *Progress in Low Temperature Physics,* edited by D. F. Brewer (North-Holland, New York, 1978), Vol. 7A; A. F. Andreev, Zh. Eksp. Teor. Fiz. **50**, 1415 (1966) [Sov. Phys. JETP **23**, 939 (1966)].

[2] See, for example, D. S. Sherrill and D. O. Edwards, Phys. Rev. B **31**, 1338 (1985).

[3] E. Krotscheck, M. Saarela, and J. L. Epstein, Phys. Rev. B **38**, 111 (1988), and references therein; N. Pavloff and J. Treiner, J. Low Temp. Phys. (to be published).

[4] B. K. Bhattacharyya, M. J. DiPirro, and F. M. Gasparini, Phys. Rev. B **30**, 5029 (1984).

[5] Sherrill and Edwards (Ref. [2]).

[6] The NMR techniques are as described in J. M. Valles, Jr., R. H. Higley, R. B. Johnson, and R. B. Hallock, Phys. Rev. Lett. **60**, 428 (1988). In an effort to observe the possible influence of substrate heating due to rf pulses on the measured values of T_1 for the work reported here, we conducted measurements of T_1 as a function of pulse energy. An increase in pulse energy was achieved by the use of the same series of rf-pulse sequences used for our reported T_1 measurements but with increased pulse length of the first rf pulse in the series. These measurements were done at 30 and 40 mK for several ^{4}He coverages. We found that an increase in pulse energy by a factor of 4 (10) yielded a 3% (30%) change in T_1. These observations convince us that for the time scale of T_1 and for the pulse energy used in our measurements, there is no significant heating effect on the T_1 values presented here.

[7] R. H. Higley, D. T. Sprague, and R. B. Hallock, Phys. Rev. Lett. **63**, 2570 (1989); in *Proceedings of the Nineteenth International Conference on Low Temperature Physics,* edited by D. S. Betts (North-Holland, Am-

sterdam, 1990), Vol. I, p. 689; Physica (Amsterdam) **165B**, 689 (1990); R. H. Higley, Ph.D. dissertation, University of Massachusetts, 1991 (unpublished).

[8] D. T. Sprague, N. Alikacem, and R. B. Hallock, in *Proceedings of the Nineteenth International Conference on Low Temperature Physics* (Ref. [7]), Vol. III, p. 539; Physica (Amsterdam) **169B**, 539 (1991); Phys. Rev. B (to be published).

[9] Nuclepore Corporation, Pleasanton, CA.

[10] Our design is patterned after that of D. S. Greywall and P. A. Bush, J. Low Temp. Phys. **46**, 451 (1982).

[11] This is a reasonable assumption since, as Fig. 1 shows. W_{LT} continues to be a linear function of $1/\sqrt{T}$ for $T > T_F$. For $T \geq 0.3$ K, the additional relaxation rate, W_{HT}, starts to dominate.

[12] C. P. Lusher, M. F. Secca, and M. G. Richards, J. Low Temp. Phys. **72**, 71 (1988).

[13] M. Himbert and J. Dupont-Roc, J. Low Temp. Phys. **76**, 435 (1989).

[14] At high temperature $M(T)$ is Curie-like, and therefore proportional to the density of ^{3}He in the film. Since 98% of the volume [6] in the cell is outside of the NMR pick-up coil, there is no contribution to the magnetization signal from the ^{3}He which is in the vapor phase, and so the magnetization is proportional to the evaporation of ^{3}He out of the film. Using the surface-to-volume ratio in the cell, we equate the chemical potential of both phases to deduce the binding energy of the film. At these high temperatures, the chemical potential of both phases was assumed to be that of an ideal gas.

[15] A. Abragam, *The Principles of Nuclear Magnetism* (Oxford Univ. Press, Oxford, 1961); W. J. Mullin, D. J. Creswell, and B. P. Cowan, J. Low Temp. Phys. **25**, 247 (1976); B. P. Cowan, J. Phys. C **13**, 4575 (1980).

[16] This assumption is supported by the fact that the decay of the spin-echo height with τ is described by a single exponential.

[17] This was solved numerically, similar to the calculation done in R. A. Guyer, K. R. McCall, and D. T. Sprague, Phys. Rev. B **40**, 7417 (1989).

[18] At the temperatures discussed here, $n_0 \sim 1$.

[19] It is perhaps reasonable to choose a power of T, such as T^2, as expected for a bulk Fermi liquid, or $\sqrt{T}$, as observed for the case of $W_0(T)$, for $W_1(T)$. We found that such a choice changes Δ by only $\sim 10\%$ and does not change significantly the quality of the exponential fit to data such as that shown in Fig. 2.

[20] $W_1 \gg W_0$ is a surprising result since one might anticipate that the two rates would be caused by similar mechanisms and therefore should be of the same order of magnitude.

[21] Given the relatively limited range of the exponential behavior, we have used various techniques to fit the data. Regardless of the technique, we find no shifts of Δ larger than the error bars; thus, we are confident that the values of Δ represent the data.

[22] J. L. Epstein and E. Krotscheck (private communication).