PHYSICAL REVIEW B VOLUME 44, NUMBER 17 1 NOVEMBER 1991-I

Longitudinal NMR relaxation of degenerate two-dimensional ^3He in ^4He films

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(Received 7 March 1991)

The NMR spin-lattice relaxation time T_1 of a degenerate submonolayer coverage of ^3He in a thin adsorbed ^4He film has been measured. As a function of ^4He coverage, T_1 exhibits a maximum and undergoes a change in temperature dependence at a coverage near that of the superfluid transition in the underlying ^4He film. Two relaxation mechanisms contribute to the observed T_1.

The ^3He-^4He mixture film constitutes a remarkable physical system. At low temperatures and submonolayer ^3He coverages, ^3He is known to reside in a bound state near the surface of ^4He forming a quasi-two-dimensional (2D) system[1-4] that has a Fermi temperature T_F which depends on ^3He coverage. The ability to tune T_F allows study of the ^3He in both classical and Fermi regimes. The system becomes more complex with increasing ^3He coverage due to ^3He-^3He interactions; rich structure is seen in the evolution to multilayer coverages of ^3He.[5-7] Recent theories are capable of predicting details of the characteristics of submonolayer coverages of ^3He interacting with the supporting ^4He film,[8-10] such as the magnetic susceptibility[11] and the specific heat;[12] these predictions are in general agreement with existing experiments. Although there has been progress in the understanding of these thermodynamic properties, much less attention has been directed towards an understanding of the dynamical properties of the mixture film.

We have carried out NMR measurements of the ^3He spin-lattice relaxation time T_1 of a submonolayer coverage of ^3He in a ^4He film as a function of ^4He coverage, n_4, for $0.20 < n_4 < 0.50$ Å^{-2}, and temperature T for $T < 300$ mK. ^4He coverages are reported as an areal density, n_4 (atoms/Å^2), and as equivalent layers of ^4He at bulk density, $D_4 = n_4 \times (3.6\ \text{Å})^2$. ^3He coverages are similarly defined, with $D_3 = n_3 \times (3.9\ \text{Å})^2$. The superfluid density is believed to be zero at the lowest ^4He coverages studied. The superfluid transition for pure ^4He is seen as a function of n_4 and T by the observation of the vanishing of third sound[13] as determined from measurements done previously;[14] for example, at $T = 100$ mK third sound is seen to vanish at a coverage $n_{4s} \simeq 0.22$ Å^{-2}. At the fixed ^3He coverage $n_3 = 6.6 \times 10^{-3}$ Å^{-2} (0.1 layers) used here, the ^3He is known to behave as a 2D weakly interacting Fermi liquid.[1,3,6] We find that T_1 displays unexpected temperature and ^4He coverage dependence. Two independent relaxation rates are observed, one of which seems correlated with the rise of the superfluid density per unit area as n_4 is increased.

The mixture films are adsorbed to a Nuclepore substrate having a surface area of 1.77 m$^2 \pm 10\%$ within the $\simeq 1$-cm^3 NMR pickup coil. Temperatures in the range 30 ± 2 mK $\leq T \leq 400 \pm 7$ mK are produced by a dilution refrigerator, and measured using a carbon resistor previously calibrated against a ^3He melting curve thermometer. Further details of the apparatus appear elsewhere.[3] Pulsed NMR techniques[15] in a 2-T magnetic field at 62.9 MHz are used to determine T_1. A series of $\pi/2 - \tau - \pi/2 - \tau_0 - \pi$ rf-pulse sequences is used to measure the relaxation of the ^3He spins to the lattice temperature as a function of time τ, with τ ranging from $\sim 0.1 \times T_1$ to $10 \times T_1$. $\tau_0 = 0.3$ msec was used for all T_1 measurements. For the coverages and temperatures reported here, T_1 was found to be well described by a single exponential over one to two decades in echo height. The magnetization and T_2, to be reported in detail elsewhere,[16] were measured using a series of $\pi/2 - \tau - \pi$ pulse sequences.

Figure 1(a) shows T_1 as a function of n_4 at various tem-

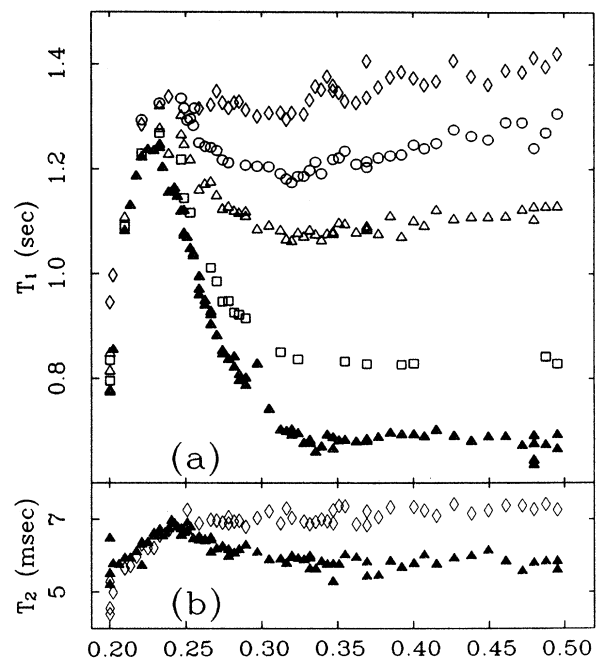

FIG. 1. (a) T_1 vs ^4He coverage for several temperatures: 30 mK (solid triangles), 50 mK (squares), 100 mK (open triangles), 150 mK (circles), and 250 mK (diamonds). T_1 (30 mK) peaks at $n_4^* = 0.23$ Å^{-2}. T_1 is nearly temperature independent for $n_4 < n_4^*$, but shows strong temperature dependence for $n_4 > n_4^*$. (b) T_2 vs n_4 for two temperatures: 30 mK (solid triangles) and 250 mK (diamonds).

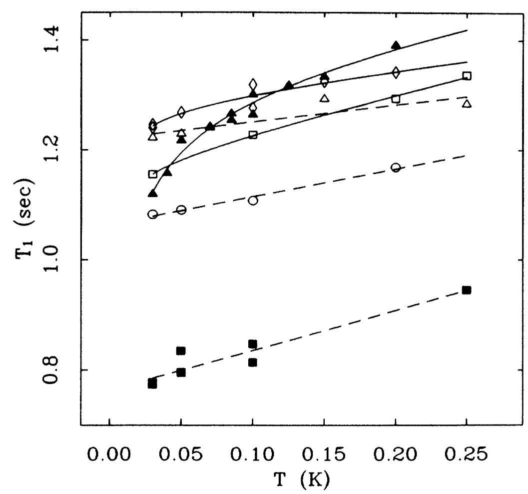

FIG. 2. T_1 vs T for various n_4. For $n_4 < n_4^*$, T_1 is weakly linear in T; at $n_4 \simeq n_4^*$ we see the onset of a stronger temperature dependence. The dashed lines are straight-line fits, while the smooth curves for $n_4 \gtrsim n_4^*$ combine the two observed temperature dependences described in the text, $T_1^{-1} = A/(1+\gamma T) + B/\sqrt{T}$. Coverages: $n_4 = 0.201$ (solid squares), 0.210 (circles), 0.222 (open triangles), 0.233 (diamonds), 0.239 (open squares), 0.248 (solid triangles).

peratures, 30 mK$\leq T \leq$250 mK. A distinct peak of width less than one layer of ^{4}He is seen in the 30-mK isotherm of T_1 centered at $n_4 = 0.23$ Å$^{-2} \equiv n_4^* \simeq n_{4s}$. No corresponding peaked structure is seen in the coverage dependence of the magnetization.[16] For $n_4 < n_4^*$, T_1 has a weak linear dependence on temperature, $T_1 \simeq (1+\gamma T)/A$, $\gamma \sim 1$ K^{-1} (Fig. 2). For $n_4 \simeq n_4^*$, T_1 is seen to deviate from linearity in T, and for $n_4 > n_4^*$ the temperature dependence of T_1 is dramatically different; $1/T_1 \simeq A + B/\sqrt{T}$ (Fig. 3). Similar $\sqrt{T}$ dependence of T_1 seen in films near 1 K (Ref. 17) has been attributed to the temperature dependence of the thermal velocity of *classical* ^{3}He. Here, the $\sqrt{T}$ behavior of T_1 is preserved well into the degenerate regime of the 2D ^{3}He (inset, Fig. 3).

The contributions to the relaxation rate, $1/T_1$, from these fits are shown (for $T = 30$ mK) as a function of n_4 in Fig. 4. We note that there is an apparent extension of $1/T_1$ for $n_4 < n_4^*$, where it is a weak function of temperature, to the temperature-independent part A of $1/T_1$ for $n_4 > n_4^*$ through the region of the peak. This suggests that $1/T_1$ for $n_4 < n_4^*$ and A for $n_4 > n_4^*$ are governed by the same relaxation mechanism which evolves smoothly through n_4^*. The increase of the B above zero for $n_4 \gtrsim n_4^*$ is interpreted as the onset of an *additional* relaxation mechanism near n_4^*. This leads us to speculate that $1/T_1$ is composed of two independent relaxation processes; $1/T_1 = W_A + W_B$ over the full range of n_4. W_A is a weak function of temperature and $W_B \sim B(n_4)/\sqrt{T}$, with $W_B \simeq 0$ for coverages $n_4 < n_4^*$. The weak temperature dependence of W_A, $W_A \sim A(n_4)/(1+\gamma T)$, is unambiguous only for $n_4 \leq n_4^*$, and gives the $T_1 \simeq (1+\gamma T)/A$ dependence observed for those coverages. Since, for larger coverages, this mild dependence is obscured by the stronger temperature dependence of W_B, we assume that the *form* of the temperature dependence of W_A does not change significantly for $n_4 > n_4^*$. Thus, the data were fit by $T_1^{-1} \simeq A/(1+\gamma T) + B/\sqrt{T}$ for all n_4, with A, B, and γ

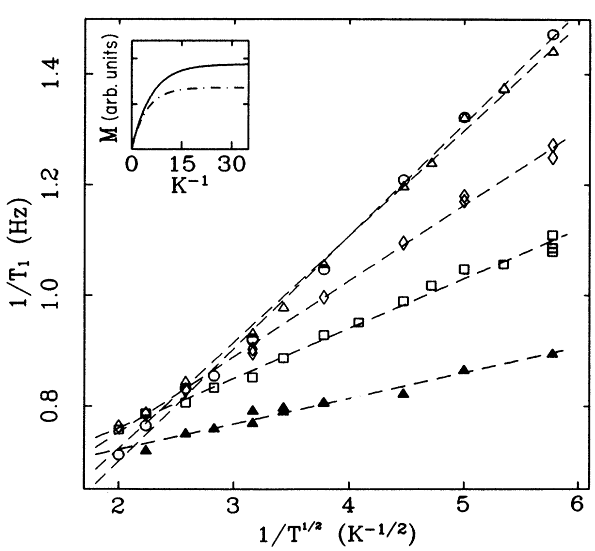

FIG. 3. T_1^{-1} vs $T^{-1/2}$ for various $n_4 \gtrsim 0.247$ Å^{-2}. The dashed lines are straight line fits to $T_1^{-1} \sim T^{-1/2}$. Coverages: $n_4 = 0.248$ (solid triangles), 0.267 (squares), 0.290 (diamonds), 0.325 (open triangles), and 0.369 (circles). Inset: Results of two-dimensional Fermi fits to the magnetization (see Ref. 16) for two representative coverages; $n_4 = 0.214 < n_4^*$ (solid curve) and $n_4 = 0.332 > n_4^*$ (dot-dashed curve), showing the spins to be degenerate for $T \lesssim 100$ mK.

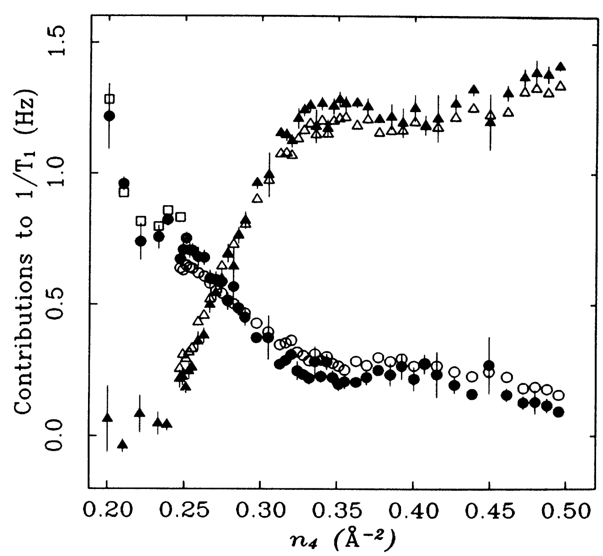

FIG. 4. Contributions to $1/T_1$ in different coverage regions from fits, shown for $T = 30$ mK. Open squares are $T_1^{-1} \sim A/(1+\gamma T)$ (valid for $n_4 < n_4^*$). Fitting to $1/T_1 \sim A + B/\sqrt{T}$ (valid for $n_4 > n_4^*$) yields A (open circles) and $B/\sqrt{T}$ (open triangles). As described in the text, the relaxation rates, W_A (solid circles) and W_B (solid triangles), are determined from a fit to $T_1^{-1} = W_A(n_4) + W_B(n_4)$, $W_A(n_4) = A(n_4)/[1+\gamma(n_4)T]$, and $W_B(n_4) = B(n_4)/\sqrt{T}$, which is valid for all coverages of n_4. These rates are shown here for $T = 30$ mK.

functions of n_4 (Table I). The effect of setting $\gamma=0$ for $n_4 > n_4^*$ does not significantly change the fit values of A and B, except near n_4^* where $\gamma \simeq B^{-2}$. The coverage dependence of these two relaxation rates W_A and W_B (at 30 mK) is shown in Fig. 4. W_A is seen to decrease relatively smoothly with increasing n_4, and is at most weakly perturbed by the onset of W_B; this supports the supposition that these are two independent relaxation mechanisms.

For dipolar relaxation due to diffusive motion in three dimensions[18] $T_1 \sim D$, where D is the spin-diffusion coefficient. Miyake and Mullin[19] have calculated that, for a degenerate 2D system, $1/D \sim T^2 \times \ln(T)$. Indeed, our preliminary measurements of the spin-diffusion coefficient at $n_4=0.34$ Å^{-2} show $D \sim T^{-2}$ (we are not able to discern the presence of the logarithmic term), but there is no evidence for $T_1 \sim T^{-2}$ in our data. In two dimensions, this simple dipolar relaxation is predicted[20] to yield a T_1 which is only weakly temperature dependent for frequencies $\omega \simeq \omega_c$, where ω_c is the spin-correlation frequency of thermal motion; this is consistent with the behavior of $W_A(T)$. The ^{3}He film is not strictly 2D, but the motion of the spins is constrained to two degrees of freedom, so the correlation functions could be expected to have the long-time tails which are responsible for the weak temperature dependence of T_1. However, our preliminary measurements of the diffusion coefficient indicate the collision frequency[21] of the ^{3}He is on the order of 4 GHz, suggesting[22] that $\omega \ll \omega_c$ for our experimental parameters. Thus, the consistency with the predictions of Ref. 20 may be fortuitous.[23]

If W_A does represent dipolar relaxation, its n_4 dependence must be explained. A coverage dependence of the effective mass, or other aspects of the mobility of ^{3}He due to the ^{4}He film, may be responsible for the coverage dependence of W_A. However, as evidenced by its temperature dependence, T_1 is not related to the spin-diffusion coefficient in a simply way. Thus, an explanation of the coverage dependence of W_A based on the effective mass seems unlikely. A dependence of the dipolar relaxation rate on the width of the ^{3}He wave function perpendicular to the substrate plane is also possible. As the wave function broadens the average strength of the dipolar interaction as a function of separation in the direction parallel to the plane of motion should decrease. Thus, the observed decrease of W_A with n_4 may be an indication of a broadening of the spatial distribution of the ^{3}He bound state with increasing n_4. Preliminary theoretical work suggests that such broadening may occur,[24] but there has been no theoretical examination of such broadening on T_1.

TABLE I. Representative fit coefficients A, γ, and B.

n_4 (Å^{-2})	A (Hz)	γ (K^{-1})	B (Hz mK$^{1/2}$)
0.200	1.246	0.785	0.011
0.210	0.978	0.614	−0.007
0.233	0.763	0.250	0.008
0.249	0.720	0.556	0.039
0.263	0.691	0.609	0.066
0.282	0.574	0.369	0.112
0.305	0.370	−0.364	0.172
0.331	0.224	−1.394	0.215
0.362	0.196	−1.491	0.220
0.427	0.188	−1.161	0.220

Next, we address the coverage dependence of W_B. For $n_4 < n_4^*$, $W_B \simeq 0$. For $n_4^* < n_4 \lesssim 0.32$ Å^{-2}, W_B increases approximately linearly with n_4 and saturates for $n_4 \gtrsim 0.33$ Å^{-2}. The range of n_4 over which this linear increase occurs corresponds to about one layer of ^{4}He. The onset of the relaxation rate W_B at n_4^* is apparently correlated with the onset of superfluidity at n_{4s}. This coverage dependence suggests a physical interpretation. If a mechanism exists whereby the underlying superfluid can facilitate the relaxation of the ^{3}He, then the strength of this interaction could depend on the amount of overlap between the ^{3}He bound surface state (of width $\sim\Lambda$) and the ^{4}He superfluid. In this scenario, the increase and saturation of W_B with an increase of n_4 is understandable. As the superfluid grows from zero with coverage at fixed temperature, the amount of superfluid sampled by the ^{3}He wave function increases; this will naturally saturate as the thickness of the superfluid blanket exceeds Λ.

We know of no mechanism involving the underlying superfluid that gives the observed $\sqrt{T}$ temperature dependence. Several naive models for this interaction prove inadequate. The superfluid film supports surface excitations such as third sound.[25] By perturbing the position of the ^{3}He atoms at the surface of the film, such excitations may cause an additional contribution to the fluctuations of the dipolar field seen by the ^{3}He. However, one would expect the magnitude of the dipolar fluctuations, and hence W_B, to be proportional to the density of those excitations, which one reasonably expects to increase with temperature, contrary to the observed temperature dependence of W_B. It is also possible that the normal cores of vortices in the superfluid may be populated by ^{3}He,[26] providing a reservoir of nondegenerate spins with which the degenerate ^{3}He could interact, a possible source of $\sqrt{T}$ behavior. However, vortices will exist in the normal state of the film as well. It is not clear to us how the binding of vortices in pairs that occurs at the K-T transition can explain the observed dramatic onset of W_B at n_{4s}.[27]

It is unlikely that the coverage or temperature dependence seen in T_1 is due to the ^{3}He relaxing with magnetic impurities on or in the surface of the Nuclepore. Since the dipolar interaction falls off with distance as $1/r^3$ one would expect surface impurities to induce motional narrowing in T_2 that is strongly ^{4}He coverage dependent. T_2 shows a temperature dependence consistent with that of T_1, but varies by no more than 15% over the entire range of coverage and temperature investigated. Thus it is unlikely that surface impurities play an important role in the relaxation of these ^{3}He submonolayer films.

In conclusion, we have studied the ^{4}He coverage and temperature dependence of the NMR relaxation times for a 0.1 monolayer coverage of ^{3}He in a ^{4}He film. We observe unexpected behavior for T_1. Theoretical work is needed to explain the observations.

We gratefully acknowledge helpful discussions with J. L. Machta and W. J. Mullin. We thank P. Sheldon for assistance with the apparatus. This work was supported by the National Science Foundation through Grant No. DMR 88-20517.

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[14]See, for example, R. H. Higley, Ph.D. dissertation, University of Massachusetts, 1991 (unpublished).

[15]In order to increase the filling factor and maximize signal strength, the NMR coil was potted directly into the epoxy sample cell, and so rf heating of the sample must be accommodated. The temperature dependence of T_1 proved useful for monitoring the equilibrium of the spin temperature, and was used to determine the time necessary to wait after rf-heating pulses. The pulse energy was chosen to be low enough so as to minimally affect the spin temperature via substrate heating on the time scale of T_1, and $\simeq 12$ min ($\sim 1000 \times T_1$) were allowed between pulse sequences to ensure thermal equilibrium of the substrate.

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[21]We take the collision frequency to be $f_c \sim k_B T_F / m^* D$. Experimentally, we find the diffusion coefficient to be $\simeq 10^{-3}$ cm^2/sec at 70 mK; the magnetization data give $T_F = 220$ mK. From Ref. 1 we estimate an effective mass, $m^*/m \simeq 1.5$.

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[23]The unique behavior predicted for T_1 in Ref. 20 results from restricting the motion of ^{3}He to a plane with a fixed orientation to the external applied magnetic field. This theory can also be applied to porous material such as Nuclepore, which present a range of such surface orientations, if the T_1 decay takes place on a short time scale compared to that of a ^{3}He moving from regions of one surface orientation to another (for more details the reader is referred to Ref. 20). Our preliminary diffusion measurements suggest that application of this so-called "powder average" is not appropriate, since the ^{3}He are able to travel an average distance of ~ 300 μm during a single T_1 decay, which is many times the average pore diameter (2000 Å) found in our Nuclepore substrate.

[24]J. Epstein and E. Krotscheck (private communication); J. Treiner (private communication).

[25]Third sound is a wavelike displacement of the ^{4}He surface. A longitudinal displacement of the ^{3}He called "surface sound" might arise [J. R. Eckardt, D. O. Edwards, P. P. Futourous, F. M. Gasparini, and S. Y. Shen, Phys. Rev. Lett. **32**, 706 (1974)]. Surface sound has not yet been observed in *thin* films. The superfluid is estimated to be too thin, ~ 5 Å, to support ripplons at 60 MHz.

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[27]With an increase of n_4, as the film becomes superfluid, the correlation function of the velocity fluctuations of the film crosses over from an exponential to a power-law decay; bound pairs of vortices exist in the film. In the normal state of the ^{4}He film, unbound (free) vortices exist. See V. Ambegaokar, B. I. Halperin, D. R. Nelson, and E. D. Siggia, Phys. Rev. B **21**, 1806 (1980).