# NMR Observation of Steps in the Magnetization of $^3$He in Thin $^3$He-$^4$He Mixture Films

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Nuclear-magnetic-resonance measurements of the magnetization of $^3$He on thin $^3$He-$^4$He mixture films show discrete structure as a function of $^3$He coverage over the range $0.0055 \leq d_3 \leq 4$ layers at a $^4$He coverage of 44 $\mu$mol/m$^2$ for $30 \leq T \leq 250$ mK in a weakly polarizing 2-T field. A steplike doubling of the magnetization at $d_3 \approx 0.8$ layer and a second less pronounced step at $d_3 \approx 1.5$ layers are ascribed to the population of higher energy levels which evolve as the $^3$He thickens. The magnetization as a function of temperature near the first step is fitted by a two-level model with an energy gap $e_{12} \approx 1.8$ K at the step.

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$^3$He adsorbed on a substrate may adopt a variety of configurations. These depend on the nature of the substrate, the temperature, and the $^3$He coverage. For example, $^3$He adsorbed on a bare substrate such as Grafoil exhibits a complicated phase diagram for submonolayer coverages, and a monolayer is solid.[1] For temperatures near 1 mK, peaks in the magnetization[2] and heat capacity[3] near second-layer completion are seen. At dilution-refrigerator temperatures, the solid causes a boundary-layer enhancement of the magnetization of overlying liquid.[4] Creation of a mixture film by the addition of a layer or two of $^4$He adjacent to the substrate renders the $^3$He a liquid and these effects disappear.[5] The $^3$He, with its larger zero-point motion in the van der Waals potential of the underlying substrate, for the most part resides on top of a thin $^4$He film, and is free to move along its nearly equipotential surface. Such $^3$He-$^4$He mixture films are an excellent system for the investigation of the $^3$He as a Fermi liquid. Measurements of the liquid are not obscured by the presence of solid $^3$He, and in the strong gradient of the van der Waals potential of the substrate one may expect to see rich behavior in the temperature and coverage dependence of the $^3$He as it evolves from two to three dimensions. The structure of mixture films and the Fermi-liquid properties of the $^3$He in them have been studied on various substrates by NMR,[6,7] heat-capacity,[8,9] torsional-oscillator,[10] and third-sound[11,12] techniques.

In this work,[13] we present NMR measurements of the magnetization of $^3$He on a thin superfluid $^4$He film. We observe the $^3$He to evolve from a two-dimensional Fermi liquid to a bulklike liquid by increasing the coverage, $d_3$, of $^3$He from 0.0055 to 4 layers at a fixed $^4$He coverage of 44 $\mu$mol/m$^2$. [Used as a unit of $d_3$, one layer is defined to be 1 atom/(3.93 Å)$^2$. Using *in situ* third-sound techniques,[7] the $^4$He film thickness is estimated to be 7.7 Å.] These measurements, made in a weakly polarizing external field of $H_0 = 2$ T over the temperature range $30 \leq T \leq 250$ mK, show two steps in the magnetization versus $d_3$ and a higher-coverage linear increase of magnetization which has the slope expected for bulk $^3$He. We interpret the stepped structure as evidence for the occupation of $^3$He states of higher discrete energy levels in the film, and discuss the first step in magnetization in terms of a two-level 2D Fermi-liquid model.

In the presentation of our measurements, we view the $^3$He film to be a Fermi liquid with quasiparticles that are free to occupy the free-particle momentum states parallel to the substrate surface, and the stationary states of the $^3$He wave functions perpendicular to the substrate surface. Hence, the energy spectrum of states is continuous in two degrees of freedom and discrete in the third. These discrete energy levels are expected to depend on the $^4$He film thickness, the van der Waals potential of the substrate, and the $^3$He coverage, and so change as $^3$He is added to the film. As we shall demonstrate, this view of the film as a finite number of interaction 2D Fermi systems is consistent with our magnetization measurements. Although our data do not determine the average position of a $^3$He particle in the mixture film, calculations[14] of the wave functions of states of a *single* $^3$He on a $^4$He film suggest that for the thin $^4$He coverage chosen for this experiment, the wave functions of the excited states lie beyond 5 Å from the substrate; thus, it is reasonable to assume that the $^3$He lies for the most part outside of the $^4$He film.

The NMR apparatus and substrate used for this experiment are as described previously.[7] The polycarbonate hydrophillic Nuclepore substrate has a van der Waals constant of $(1.9 \pm 0.5) \times 10^{-21}$ cm$^3$K.[15] Its 0.2-$\mu$m-diam pores are not capillary condensed[16] at the coverage used in this experiment, and provide most of the 1.77 m$^2 \pm 10\%$ surface area within the sample cell.

The magnetization $M$, defined in terms of the entire sample of $N$ spins each of magnetic moment $\mu_m$ by $M \equiv \mu_m(N_+ - N_-)$, was measured by use of 90°-$\tau$-180° spin-echo pulse sequences. Echo heights measured for several values of $\tau$ extrapolated to $\tau = 0$ provide a measure of the magnetization, which is calibrated to an uncertainty of 6% by Curie-law data from the lowest coverages. Most of the magnetization and NMR $T_2$ data was determined by 3–4 echoes for $0.3 \leq \tau \leq 1.5$ msec. The

 

$T_1$ and $T_2$ relaxation mechanisms for submonolayer coverages of $^3$He in mixture films on this substrate are not well understood,[17] and so consequently we focus on the magnetization measurements, although we will comment on the $T_2$ data later.

The data are normalized by $M_0 = N\mu_m^2 H_0/k_B T_F(m_3)$, the magnetization at $T=0$ of an area $A$ of a weakly polarized ($\mu_m H_0 \ll k_B T_F$) 2D ideal Fermi gas (2DIFG) of $N$ particles each with mass equal to the bare mass, $m_3$, of a $^3$He atom. Note that in two dimensions the Fermi temperature is $T_F(m) = \pi\hbar^2 N/k_B m A$, and thus $T_F \propto N$ and $M_0$ is independent of $^3$He coverage.

A portion of the $0.0055 \le d_3 \le 0.85$ layer magnetization data is shown in Fig. 1. In the low-coverage data, $d_3 < 0.3$ layer, where $dM/dT < 0$, the Pauli paramagnetism of a 2D Fermi liquid is seen. To illustrate, we show in Fig. 1 fits to these low-coverage data by the expression for the magnetism of a weakly polarized 2DIFG,

$$M_m(T)/M_0 = (m/m_3)\{1 - \exp[-T_F(m)/T]\}, \quad (1)$$

using the quasiparticle mass $m$ as a fitting parameter at each coverage.[18] An additional fitting parameter for the $d_3 \le 0.022$-layer data, where the Curie-law behavior is most prominent, determines the calibration constant.

As the $^3$He coverage increases, quasiparticle-quasiparticle interactions are expected. The magnetization at $T=0$ of a submonolayer of $^3$He may be discussed in terms of 2D Landau Fermi-liquid theory as[19]

$$M(T=0)/M_0 = (m^*/m_3)/(1+F_0^a),$$

where $F_0^a$ is a Fermi-liquid parameter for antisymmetric interactions, and $m^*$ is the effective mass, which determifnes the 2D density of states $g(m^*) = m^* A/\pi\hbar^2$.

Figure 2 shows a plot of $M/M_0$ vs $d_3$ at $T=40$ mK, the lowest temperature for which there are data at each coverage. For $0.1 \le d_3 \le 0.65$ layer the magnetization is degenerate at $T=40$ mK. Over this range the linear increase with $d_3$ of $M$(40 mK) is ascribed to density-dependent interactions among the $^3$He,[7] and a linear extrapolation to $d_3=0$, where there is no quasiparticle-quasiparticle interaction, gives the hydrodynamic mass $m_h = 1.38 m_3 \pm 16\%$. By a variational technique, Krotscheck, Saarela, and Epstein[20] have calculated $M(T=0)/m_h M_0$ for a range of $d_4$ and $d_3 \le 0.5$ layer. For $d_4 = 44$ $\mu$mol/m$^2$, they find a linear increase of $M(T=0)/m_h M_0$ with $d_3$ up to $d_3 \approx 0.25$ layer with slope approximately 2.36 layer$^{-1}$, which may be compared to the value $1.9 \pm 0.3$ layer$^{-1}$ obtained from the data shown in Fig. 2.

From $d_3 = 0.7$–$0.9$ layer, above the lowest coverage for which $dM/dT > 0$ in Fig. 1, a steplike doubling of $M$(40 mK) occurs. A second step is less prominent at $d_3 \approx 1.5$ layers. These steps in $M$(40 mK) suggest occupation of higher discrete energy levels in the film by $^3$He atoms; the positive $dM/dT$ preceding the first step suggests thermal excitation into the second level. Once the third level is partially populated, the magnetization increases at the same rate per particle as would bulk $^3$He, within the uncertainty in the Curie-law calibration, and no fur-

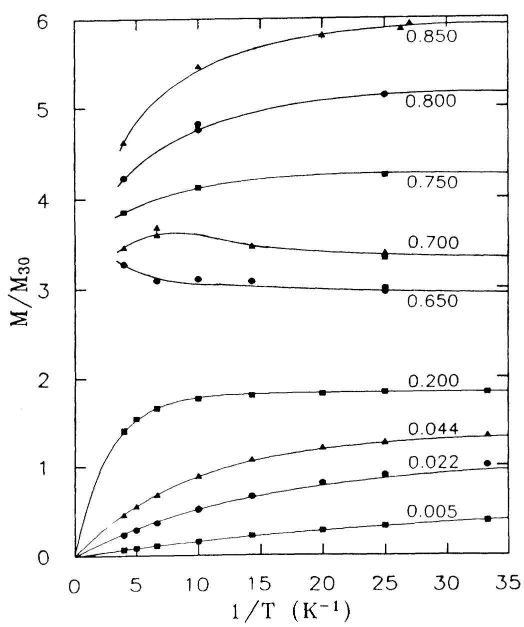


FIG. 1. Magnetization of the sample vs $1/T$ at fixed $^4$He coverage 44 $\mu$mol/m$^2$ for the $^3$He coverages shown at the right. The positive $dM/dT$ at $d_3 = 0.65$ layer precedes the step in magnetization seen in Fig. 2. The curves for $d_3 \le 0.2$ layer are 2D Fermi-gas fits, and the curves for $d_3 \ge 0.65$ layer are a guide to the eye.

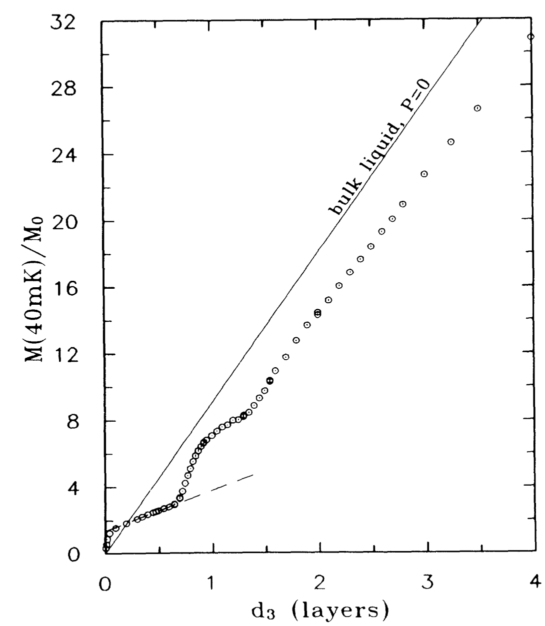


FIG. 2. Magnetization at $T=40$ mK vs $d_3$, showing steps in magnetization at $d_3 \approx 0.8$ and 1.5 layers, and an increase in magnetization with the same slope as for bulk liquid above $d_3 = 2$ layers. The dashed line extrapolates low-coverage data to obtain the hydrodynamic mass $m_h = 1.38 m_3$.

ther steps in magnetization are seen. Additional $^3$He beyond completion of the second level may be considered to increase the number of the $^3$He atoms having neighbors on all sides, thus having nearly bulklike interactions and magnetization.

To illustrate the interpretation of the steps as population of higher energy levels, we apply a simple two-level model to the data in the vicinity of the step at $d_3 \approx 0.8$ layer. We assume for this model that the magnetization may be calculated by summing the magnetization of two levels of 2D Fermi liquid composed of quasiparticles in different states in the mixture film having discrete energy levels $e_1$ and $e_2$ in the vicinity of the step. At low temperature, quasiparticles added below $d_3 \approx 0.8$ layer go into the first level. When the chemical potential $\mu$ reaches $e_2$, quasiparticles begin to populate the second level, thus creating a step increase in the magnetization. Because the magnetization approximately doubles at the step, we assume that all the quasiparticles share the same $m^*$ and $F_0^a$.

The step in magnetization near $d_3 = 0.8$ layer shown in Fig. 2 has a width in $d_3$ which is greater than expected from thermal excitation at $T = 40$ mK. Indeed, if the $M(T)$ data at each coverage are extrapolated to $T = 0$, a plot of the extrapolated $M(T=0)$ vs $d_3$ shows a substantial width to the step, even after allowing for a generous uncertainty in the extrapolations. Therefore, we assume that substrate inhomogeneities[18] contribute to the width of the step, and the model is allowed a fit parameter, $\sigma$, for the width of a Gaussian distribution in $e_{12}$.

In lieu of a theory for the temperature dependence of the magnetization of a dense single-level 2D Fermi liquid, we use an approximation of Havens-Sacco and

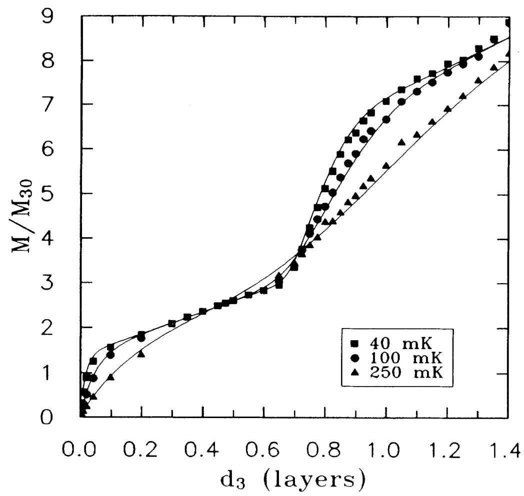


FIG. 3. Magnetization vs $d_3$ at 40, 100, and 250 mK. Also shown is a fit by a two-level model having energy gap $e_{12} \approx 1.8$ K near the step at $d_3 = 0.74$ layer and a distribution in $e_{12}$ of $\sigma \approx 0.10$ K. We note that $dM/dT \approx 0$ at $d_3 = 0.71$.

Widom[19] valid in the dilute limit,

$$M(T)/M_0 = \frac{M_{m_h}(T)}{1 - F M_{m_h}(T)}, \tag{2}$$

where $M_{m_h}$ is the magnetization of a 2DIFG of particles of mass $m_h$ as given by Eq. (1), and the effect of interactions is contained in the function $F$. In this formulation, only spherical-wave quasiparticle-quasiparticle interactions are considered; thus $m^* = m_h$, and the density of states is $g = m_h A/\pi\hbar^2$. For a two-level model, we let the chemical potential determine the number of particles in each level, and write

$$\frac{M(T)}{M_0} = \frac{\chi_1(T)}{1 - F\chi_1(T)} + \frac{1}{(2\pi)^{1/2}\sigma}\int_{-\infty}^{\infty} du\, e^{(e_{12}-u)^2/2\sigma^2}\frac{\chi_2(T)}{1 - F\chi_2(T)}, \tag{3}$$

where $\chi_1 \equiv (m_h/m_3)/(1 + e^{-\mu/k_B T})$ and $\chi_2 \equiv (m_h/m_3)/(1 + e^{(u-\mu)/k_B T})$ give 2DIFG magnetizations corresponding to $M_{m_h}$ in terms of $\mu$. $F$ as a function of $d_3$ is determined by a linear fit to degenerate plateaus in $M$(40 mK)$/M_0$ vs $d_3$ below and above the step, since in the degenerate limit of this model

$$M(T=0)/M_0 = (m_h/m_3)/[1 - F(d_3)]$$

well below the step, and twice this quantity well above the step. The result of a fit varying the location, $d_{3S}$, of the step and $\sigma$ as fitting parameters is shown in Fig. 3 for $\sigma = 0.1$ K and $d_{3S} = 0.74$ layer (i.e., $e_{12} = d_{3S}/g = 1.8$ K).

The fit shows good agreement with the data, despite the use of the Havens-Sacco–Widom temperature dependence beyond the weak-interaction limit of its validity. The value of $e_{12}$, and the model itself, may give some insight into the structure of the film and provide a framework for discussion, but should not be considered definitive. The discrete energy levels of $^3$He near the step may be expected to depend strongly on the population of each level. For example, the assumption that $e_{12}$ is approximately constant across the step would be invalid if hard-sphere packing determines when levels fill. This would be consistent with the $\Delta d_3 \approx 0.7$-layer interval in the magnetization steps, as well as peaks seen earlier[11] in the $Q$ of third sound at $d_3 = 0.7$ and 1.3 layers on mixture films with thicker $^4$He coverages in a glass resonator.

$T_2$ is not as definitive a probe of multiple-layer coverages of liquid $^3$He on a mixture film as it is for $^3$He on a bare substrate. In contrast to the dramatic factor-of-100 reduction in $T_2$ seen at monolayer completion of $^3$He on a bare Grafoil substrate,[21] our measurements of $T_2$ at constant temperature show little dependence on $d_3$. At $T = 40$ mK, $T_2 = 7 \pm 1$ msec for all coverages, although an increase in $T_2$ of roughly 10% appears discernible at $d_3 \approx 0.7$ layer, in the vicinity of the first step in the magnetization. $T_1$ ($T_1 \approx 1$ sec) was not measured systematically across the step in magnetization. Both $T_1$ and $T_2$ increase weakly with temperature [$T_2(250\,\mathrm{mK}) = 8$ msec].

In contrast to these films' structured evolution to 3D behavior, surface-tension measurements[22] determining the entropy $S$ of $^3$He on the surface of a bulk mixture near phase separation approach the bulk slope for $S/T$ vs $d_3$ after $d_3 = 0.5$ layer without any steps. The relatively weak van der Waals potential above the bulk mixture may explain the lack of steps, which wash out to give bulklike behavior when the temperature approaches or exceeds the energy separation between what would otherwise be identifiable levels.

In conclusion, the magnetization of $^3$He on a thin $^4$He film shows discrete structure as a function of $^3$He coverage which is interpreted as the occupation of second and third energy levels in the film, followed by a linear increase of magnetization with slope as for bulk fluid. These experimental results should further stimulate theoretical work[23] on mixture films.

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