Nuclear Magnetic Susceptibility Measurements of ^{3}He-^{4}He Mixture Films

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Nuclear magnetic susceptibility measurements of the ^{3}He in dilute ^{3}He-^{4}He mixture films are presented. At low ^{3}He coverages ($d_3 < 0.1$ layer), the ^{3}He behaves approximately as a 2D ideal Fermi gas. For larger submonolayer ^{3}He coverages, the low-temperature susceptibility is enhanced, increasing with increasing amounts of ^{3}He, contrary to the 2D ideal Fermi-gas model. No evidence for a phase transition in the ^{3}He is seen.

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At low temperatures, a dilute concentration of ^{3}He "floats" on a thin film of ^{4}He and behaves as a two-dimensional Fermi fluid. This system has been extensively studied in third-sound,[1,2] torsional-oscillator,[3,4] heat-capacity,[5,6] and NMR[7] experiments. The heat-capacity measurements are of particular interest because they indicate a possible gas-to-liquidlike phase transition in the two-dimensional gas of ^{3}He quasiparticles.

In this Letter, we present NMR measurements of the ^{3}He susceptibility χ in dilute ^{3}He-^{4}He mixture films as a function of ^{3}He and ^{4}He coverage and temperature. For ^{3}He coverages d_3 less than 0.1 monolayer adsorbed on a ^{4}He film of thickness $d_4 = 2.8$ atomic layers, the susceptibility data as a function of temperature agree reasonably well with the predictions for a two-dimensional ideal Fermi gas (2DIFG). For larger submonolayer ^{3}He coverages with $d_4 = 9.5$ layers, we find an enhancement of the $T = 0$ susceptibility from the 2DIFG model which can be attributed to ^{3}He-^{3}He interactions which we discuss within a Landau-Fermi-liquid framework. The susceptibility is found to depend only weakly on ^{4}He film thickness for $2.8 \leq d_4 \leq 9.5$ layers, for $d_3 = 0.088$ layer.

The sample cell was designed for simultaneous NMR and third-sound studies of mixture films at dilution-refrigerator temperatures. The substrate consisted of Nuclepore polycarbonate filters[8] of nominal thickness 10 μm perforated by $\simeq 4\times 10^8$/cm^2 approximately cylindrical pores of 200 nm nominal diameter. These filters provided a large surface area with a well-defined and characterized geometry. Most of the sample-cell surface area $A = 1.77$ m^2 was provided by 400 such Nuclepore filters 13 mm in diameter with a 3-mm-diam hole in the center. These filters were pressed onto a 3-mm-diam copper post located on the axis of the NMR coil. The copper post was welded to the copper support structure for the sample cell which was in good thermal contact with a sintered copper plug residing in the mixing chamber of a dilution refrigerator. The temperature was measured with a Speer 100-Ω carbon resistor which was calibrated against the ^{3}He melting curve[9] to an accuracy of ± 3 mK.

Our mixture-film coverages are expressed in terms of bulk-density atomic layers. One atomic layer of ^{3}He corresponds to an areal density of 6.4×10^{14} atoms/cm^2. We express the ^{4}He coverage in terms of the distance d_4 (in 3.6-Å-thick layers) from the free surface of the ^{4}He film to the substrate. For the ^{4}He coverages of interest here, third-sound measurements in this sample cell[10] on pure ^{4}He films show d_4 to be related to the areal density of ^{4}He through $d_4 = (D_4 - 4.8\ \text{Å})/3.6\ \text{Å}$, where $D_4 = N_4/An_4$, $n_4 = (3.6\ \text{Å})^{-3}$ is the number density in bulk ^{4}He liquid, and N_4/A is the ^{4}He areal density.

The thermal-equilibrium ^{3}He magnetization M_0 was measured by pulsed NMR in a 2-T (Larmor frequency = 62.9 MHz) magnetic field H_0 oriented perpendicular to the average Nuclepore pore axis. The height $E(\tau)$ of the spin echo obtained following a 90-τ-180 pulse sequence was measured for a number of different values of τ. The extrapolation to $\tau = 0$ of $\ln E(\tau)$ provided a measure of M_0. For most[11] of the data we present here $\ln E(\tau)$ was linear in τ indicating an exponential decay of the phase correlation function of the spins characterized by a single time constant T_2 which was typically several milliseconds. To ensure that the substrate and the ^{3}He magnetization returned to their original temperature after being heated by the NMR pulses, we waited several minutes between successive 90-τ-180 pulse sequences. The magnetization of the ^{3}He was observed to be affected by rf heating only after a time of the order of the spin-lattice relaxation time T_1 (typically 1 s), whereas the echos used to measure the susceptibility occurred several milliseconds after the first rf pulse.

The susceptibility $\chi = M_0/H_0$ was obtained from the echo height $E(0)$ by our assuming $E(0) = \beta\chi H_0$, where β is a constant characterizing the spectrometer sensitivity. The value of β was determined from the Curie-type susceptibility data (all the data points with $\chi/\chi_{30} < 0.8$ in Fig. 1) with use of the expression

$$E(0) = (\beta\hbar^2\gamma^2 H_0/4k_B)[(N_3/T) + B(N_3/T)^2],$$

where N_3 is the measured number of ^{3}He atoms in the sample cell, γ is the ^{3}He gyromagnetic ratio, and β and

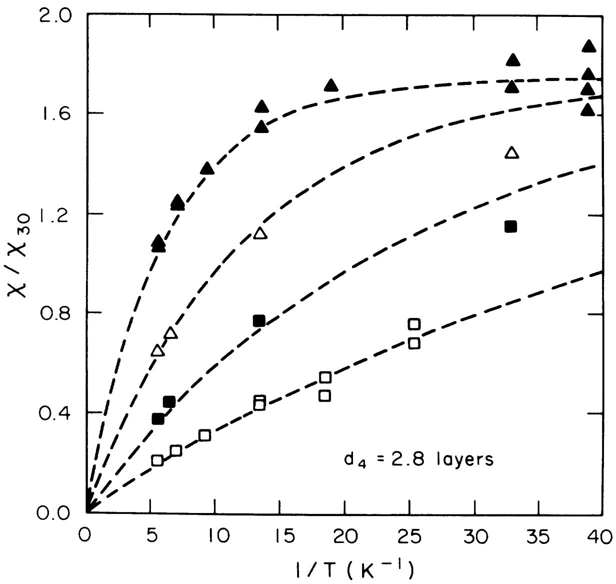

FIG. 1. Susceptibility as a function of inverse temperature for low ^{3}He coverage and $d_4=2.8$ layers. ^{3}He coverages d_3, in layers: open squares, 0.011; filled squares, 0.022; open triangles, 0.044; filled triangles, 0.088. The dotted lines are a fit to the 2DIFG model.

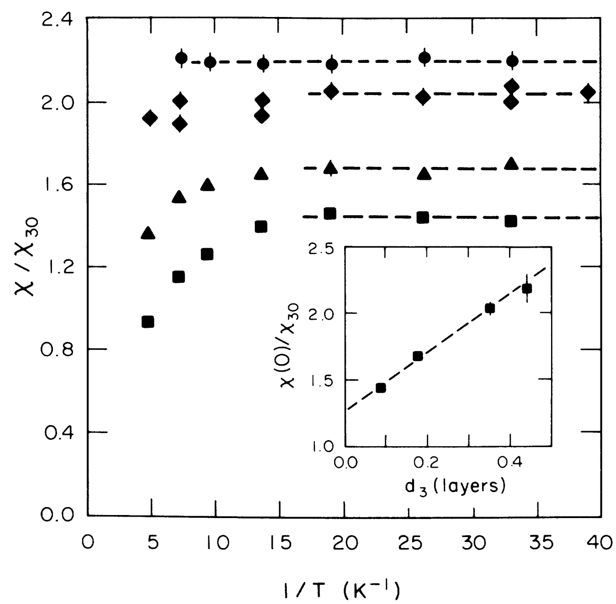

FIG. 2. Susceptibility as a function of inverse temperature for high ^{3}He coverage and $d_4=9.5$ layers. ^{3}He coverages d_3, in layers: squares, 0.088; triangles, 0.176; lozenges, 0.352; circles, 0.440. The dashed lines indicate extrapolations used to obtain the values of $\chi(0)$ which are plotted as a function of d_3 in the inset. The dashed line in the inset is a linear fit to the data.

B are fitting parameters.

We present susceptibility data as a function of inverse temperature in Figs. 1 and 2 for a range of ^{3}He coverages at two fixed ^{4}He coverages. The temperature dependence of the data is qualitatively as one would expect for a Fermi system; the susceptibility evolves smoothly from an almost linear dependence on $1/T$ for $T_F/T \ll 1$ to no dependence on the temperature for $T_F/T \gg 1$.

For the low-^{3}He-coverage data shown in Fig. 1, the data can be fitted reasonably well by $\chi(T)$ for a 2DIFG. The magnetic susceptibility as a function of temperature for a spin-$\frac{1}{2}$ 2DIFG in low field is given by

$$\chi(T)=(\gamma^2/4\pi)Am_H[1-\exp(-T_F/T)], \quad (1)$$

where $T_F=\pi\hbar^2N_3/m_Hk_BA$ and m_H is the hydrodynamic mass of an isolated ^{3}He quasiparticle. In the figures, we have normalized our data by $\chi_{30}=\gamma^2Am_3/4\pi$, where χ_{30} is the $T=0$ susceptibility of a spin-$\frac{1}{2}$ 2DIFG with mass m_3, where m_3 is the bare mass of a ^{3}He atom. For a 2DIFG, χ depends on T only through T_F/T, which is proportional to N_3/T, so that all the susceptibility data for a 2DIFG at various ^{3}He coverages and temperatures (at a constant ^{4}He coverage) can be fitted *simultaneously* with N_3/T as the independent variable, with m_H as the only fitting parameter. This analysis assumes that the areal density of ^{3}He remains uniform over the area A as N_3 is varied. The results of fitting our data with use of this procedure are shown in Fig. 1 as one smooth curve for each ^{3}He coverage, with $m_H/m_3=1.8$. This value of m_H/m_3 implies a Fermi temperature of 160 mK for $d_3=0.088$ layer. Modest deviations from the 2DIFG model may be present.

Deviations from 2DIFG behavior are clearly seen at higher ^{3}He coverages, as shown by the data in Fig. 2, taken with a thicker ^{4}He film (9.5 layers). The low-temperature value of the susceptibility (see inset of Fig. 2) monotonically increases with d_3, contrary to the behavior predicted by Eq. (1) for a 2DIFG.

It is likely that this deviation from 2DIFG behavior is due to ^{3}He-^{3}He interactions. In a Landau-Fermi-liquid theory, the susceptibility of a two-dimensional weakly interacting Fermi system is given by[12,13]

$$\chi(0)/\chi_{30}=(m_H/m_3)(1+F_1^s/2)/(1+F_0^a),$$

where only the two-dimensional Fermi-liquid parameters F_1^s and F_0^a are predicted[12,14] (at low density) to depend on the ^{3}He areal density. The density of states at the Fermi surface is calculated from the effective mass $m^*=m_H(1+F_1^s/2)$. The data show that $(1+F_1^s/2)/(1+F_0^a)$ increases approximately linearly with d_3. On the basis of this observation, we have extrapolated the best linear fit to the data in the inset of Fig. 2 to $d_3=0$, where F_1^s and F_0^a are zero; we find $m_H/m_3=1.26\pm0.15$.

The approximately linear increase of $\chi(0)$ with d_3 is consistent with earlier χ measurements by Brewer, Creswell, and Thomson[7] on ^{3}He-^{4}He mixture films adsorbed on Vycor having 60-Å-diam pores at higher temperatures and ^{3}He coverages than we have explored. They measured $\chi(T)$ for films with a fixed ^{3}He concentration of 9% for three different values of the *total* film thickness at temperatures between 0.3 and 1.5 K. In terms of our parameters, they studied $0.5 \leq d_3 \leq 1.0$ layer and $3 \leq d_4 \leq 6$ layers. Through this range of d_3, $\chi(0)$ increased approximately linearly with roughly the same slope and intercept as our data. We emphasize that our results in Fig. 2 were obtained at constant ^{4}He coverage, unlike those of Ref. 7. We have found (see below) that $\chi(0)$ depends on d_4. Hence, quantitative comparisons between our results and the results of Brewer *et al.* are difficult.

On the basis of calculations of the density profile perpendicular to the free surface of a pure ^{4}He film[15] and the binding energy of ^{3}He to the free surface of a ^{4}He film,[16] we might expect the ^{3}He quasiparticles on a 9.5-layer film to have very nearly the same properties as ^{3}He quasiparticles on the surface of bulk ^{4}He liquid. In fact, the value of $m_H/m_3 = 1.26 \pm 0.15$ obtained from the linear fit to the data in the inset of Fig. 2 ($d_4 = 9.5$ layers) is consistent with the bulk surface result[17] of 1.45 ± 0.1. However, surface tension and surface sound-velocity measurements by Edwards and co-workers[17] indicate that ^{3}He quasiparticles on the surface of bulk ^{4}He liquid behave as a nearly 2DIFG for areal densities up to $d_3 = 0.32$ layer. They found the ^{3}He-^{3}He interactions to be weak and to have a negligible effect on the d_3 dependence of the $T=0$ surface tension and surface sound velocity. Within a Landau-Fermi-liquid theory, the surface tension and the surface sound velocity would be expected to depend only on the spin-symmetric part of the Landau interaction functional. The susceptibility, on the other hand, depends on the spin-symmetric and spin-antisymmetric parts of the Landau interaction functional. Therefore, our data could be reconciled with the data of Ref. 17 if the dependence of $\chi(0)$ on d_3 is due only to the spin-antisymmetric part of the Landau interaction functional.

The ^{4}He film thickness dependence of the susceptibility at fixed d_3 is presented in Fig. 3. The qualitative temperature dependence of the susceptibility depends little on d_4, but the magnitude of $\chi(0)$ decreases as d_4 increases. We have fitted these data at each ^{4}He coverage with a function of the form $\chi(T) = \chi(0)[1 - \exp(-b/T)]$, with b and $\chi(0)$ as fitting parameters. The dependence of $\chi(0)$ on d_4 (shown in the inset in Fig. 3) can be attributed to the structure- and film-thickness-dependent short-wavelength modes of the free surface of the ^{4}He film.[15,17] These modes may affect $\chi(0)$ by influencing the ^{3}He-^{3}He interactions or the hydrodynamic mass of the ^{3}He quasiparticles.

Finally, we note that our susceptibility data at the lowest ^{3}He and ^{4}He coverages were taken through the coverage region in which ^{3}He heat-capacity measurements showed interesting deviations from 2DIFG behavior. Briefly, the ^{3}He heat capacity had the temperature dependence of a 2DIFG when $d_3 \geq 0.2$ layer and $d_4 \geq 4.0$ layers.[5] However, at smaller ^{3}He and ^{4}He coverages, the heat capacity deviated from this simple model in a manner which has been interpreted as due to a 2D puddling (gas-to-liquid) transition of the surface ^{3}He quasiparticles.[6] A clear change of slope in the heat capacity versus temperature below 150 mK indicated the transition. Given the smooth dependence of the susceptibility on temperature shown in Figs. 1 and 3, it is clear that we see no signs of a similar abrupt transition for the coverages we have studied. However, this does not necessarily exclude the possibility of a puddling transition, since some features of the heat-capacity data (e.g., the absence of a latent-heat peak) are not sufficiently well understood to predict the behavior of $\chi(T)$ near the transition. The low-temperature slope of the heat capacity $dC/dT(0)$ was observed to be proportional to N_3. This was interpreted as evidence that virtually all the ^{3}He had condensed into a high-density phase (puddles), with the density of ^{3}He in the puddles constant, and the area occupied by the puddles increasing in proportion to N_3. However, this puddle model unambiguously predicts that $\chi(0)$ is also proportional to N_3. Our lowest-temperature data in Fig. 1 clearly show that $\chi(0)$ is *not* proportional to N_3, given the physically reasonable assumption that $\chi(T)$ does not decrease at temperatures

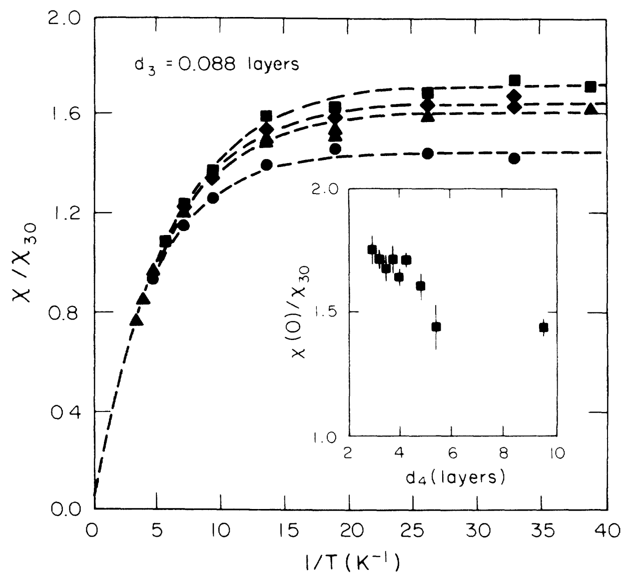

FIG. 3. Susceptibility as a function of inverse temperature for several ^{4}He coverages and $d_3 = 0.088$ layer. ^{4}He coverages d_4, in layers: squares, 3.1; lozenges, 3.9; triangles, 4.75; circles, 9.5. The dashed-line fits are described in the text. Inset: Zero-temperature susceptibility obtained from the fits to the data in the main part of the figure and data from additional ^{4}He coverages.

lower than our lowest available temperatures. The different N_3 dependences of $\chi(0)$ and $dC/dT(0)$ are unlikely to be explained by magnetic interactions in a puddling model, since these would be expected to depend only on the ^{3}He density in the condensed phase, rather than on N_3.

In summary, we have used pulsed NMR techniques to measure the susceptibility of ^{3}He in dilute ^{3}He-^{4}He mixture films for a range of ^{3}He and ^{4}He coverages at temperatures between 30 and 180 mK. We find that at low ^{3}He coverages, the susceptibility is approximately that of a 2D ideal Fermi gas (2DIFG), but at higher ^{3}He coverages, the susceptibility is enhanced compared with that of a 2DIFG. We attribute this enhancement to the influence of ^{3}He-^{3}He interactions. Further work is needed (1) on 2D Landau-Fermi-liquid theory[18] and (2) to obtain a model consistent with both the heat capacity and susceptibility data at low temperatures.

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