

## Quasiparticle Interactions in Two-Dimensional $^3\text{He}$ on $^4\text{He}$ Films

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Measurements are reported of the heat capacity of  $^3\text{He}$  bound to an atomically layered  $^4\text{He}$  film on the surface of graphite. At  $^3\text{He}$  coverages  $n_3 < 4 \text{ nm}^{-2}$ , the  $^3\text{He}$  forms a uniform 2D Fermi system. Thereafter with increasing  $n_3$  we resolve two steps in the heat capacity, each arising from the formation of a further 2D continuum built on excited surface-normal states. We infer information on the hydrodynamic mass, quasiparticle interactions, and energetics of these states. Both short range repulsive interactions and ripplon mediated quasiparticle interactions are important. [S0031-9007(99)09131-0]

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The surfaces of bulk liquid  $^4\text{He}$  and adsorbed  $^4\text{He}$  films provide ideal substrates for the study of two-dimensional  $^3\text{He}$ .  $^3\text{He}$  atoms are bound to the surface of bulk liquid  $^4\text{He}$  with an energy of  $-5 \text{ K}$ , compared to the bulk binding energy of  $-2.8 \text{ K}$ , and form a weakly correlated two-dimensional Fermi system [1]. The existence of such a bound state (Andreev state [2]) first became apparent in surface tension measurements. The inferred hydrodynamic effective mass in this state, arising from coupling to the excitations of the  $^4\text{He}$  surface (ripples), is  $m_H/m = 1.45 \pm 0.1$ . It was originally thought that at  $^3\text{He}$  coverages above  $6.4 \text{ nm}^{-2}$  further atoms would simply dissolve in bulk. However, a second surface bound state has been proposed theoretically whose existence depends strongly on the surface profile of the film [3]. More recently there has been intense interest in adsorbed  $^3\text{He}$ - $^4\text{He}$  mixture films, for which several surface-normal bound states are predicted for the  $^3\text{He}$  impurity [4]. Our interest is in the 2D Fermi liquids that can be built on these states. Previous experimental studies of helium mixture films, mostly using the heterogenous substrate Nuclepore filter paper, include an extensive series of heat capacity measurements for  $T > 40 \text{ mK}$  [5] and NMR studies at  $T > 25 \text{ mK}$  [6–9], which clearly support the existence of two surface-normal states.

In these 2D Fermi fluids, quasiparticle interactions are expected via ripplon mediated interactions as well as direct  $^3\text{He}$ - $^3\text{He}$  interactions. The versatility of the helium films lies in the fact that the interactions can be tuned both by varying the  $^4\text{He}$  film thickness and by changing the  $^3\text{He}$  coverage. Understanding such interactions is important for the consideration of potential instabilities of the Fermi fluid state, such as superfluidity [10,11], dimerization [12], and 2D condensation.

In this paper we report the first heat capacity study of  $^3\text{He}$  adsorbed at the surface of a superfluid  $^4\text{He}$  film which is in turn adsorbed on the atomically flat surface of graphite. Our use of a graphite substrate has been motivated by the fact that, due to its homogeneity, the helium films exhibit layer by layer growth [13]. This is particularly important for producing well characterized thin  $^4\text{He}$  films. In addition, films on graphite can readily be cooled

to submillikelvin temperatures [14]. The present heat capacity measurements extend to  $1 \text{ mK}$ , deep into the fully degenerate regime.

There is a significant body of theoretical work on such films. Predictions have been made as a function of  $^4\text{He}$  fluid coverage for a composite substrate consisting of graphite plus two solid layers [15], and this provides additional motivation for the present experimental study of this system. First-principles calculations of the hydrodynamic effective mass of the  $^3\text{He}$  impurity state adsorbed on the film show interesting structure as a function of  $^4\text{He}$  coverage due to atomic layering of the film [15–17]. In addition, the energetics of  $^3\text{He}$  states in the film have received considerable attention. Studies of  $^3\text{He}$  quasiparticle interactions in these systems are less advanced [18].

The main conclusions of the present experiments are as follows. We find that the 2D Fermi fluid built on the surface-normal ground state is stable with respect to 2D condensation, in contrast to the conclusion from previous heat capacity measurements on helium mixture films [19]. By varying the coverage of the  $^3\text{He}$  layer, we can distinguish the hydrodynamic and interaction contributions to the effective mass. Above some  $^3\text{He}$  coverage, population of the first excited surface-normal state gives rise to a step in the heat capacity, with a width consistent with thermal broadening. The location of this step at a coverage significantly smaller than one  $^3\text{He}$  “layer” ( $6.4 \text{ nm}^{-2}$ ) provides evidence for ripplon mediated interactions in the film. Since our measurements extend to low millikelvin temperatures, we are able to directly infer the hydrodynamic mass of this second state, which appears to overlap more strongly with the  $^4\text{He}$  film than the ground state, as predicted theoretically [15,17]. A subsequent step suggests the existence of a further bound state, as predicted theoretically [15,17] and unobserved hitherto.

In this work the  $^4\text{He}$  film consisted of four atomic layers, comprising two solid layers plus two superfluid layers, corresponding to a coverage  $33.5 \text{ nm}^{-2}$ . Layer promotions are identified by successive compressibility minima inferred from vapor pressure isotherms, made at  $940 \text{ mK}$  using an *in situ* pressure gauge, which allows

the preparation of such a film with an integral number of atomic layers with good precision. The estimated fluid coverage is  $13 \text{ nm}^{-2}$ . The heat capacity measurements were performed on the same cell as used in previous work [20]. The exfoliated graphite substrate has a surface area of [21]  $182 \text{ m}^2$ . We measure the background heat capacity of the empty cell and assume the heat capacity of the  $^4\text{He}$  film to be negligible in this temperature regime [5].

The heat capacity data are shown, after background subtraction, for a selection of  $^3\text{He}$  coverages in Fig. 1. The low temperature heat capacity is linear in temperature as expected for a degenerate Fermi fluid. There is no temperature independent contribution to the heat capacity, such as observed in pure  $^3\text{He}$  films on bare graphite [22] and HD plated graphite [23], and usually attributed to  $^3\text{He}$  atoms localized by residual substrate heterogeneities.

Above a coverage  $n_s = 4.2 \text{ nm}^{-2}$  heat capacity isotherms, Fig. 2, show a steplike increase in the heat capacity that arises from population of the first excited surface-normal state. In the following we discuss the coverage regimes  $n_3 < n_s$  and  $n_3 > n_s$  in turn.

For  $n_3 < n_s$  the system consists of a 2D Fermi fluid of uniform density. This follows since the measured linear coefficient of the heat capacity  $\gamma = c/T = (\pi^2/3)k_B^2 g = \pi k_B^2 m^* A / 3\hbar^2$  is only weakly dependent on surface density,  $n_3$ , over the whole coverage range. Here  $g = Am^*/\pi\hbar^2$  is the energy-independent density of states and  $m^*$  is the  $^3\text{He}$  effective mass. The Fermi temperature of such a 2D gas is  $T_F = 0.505n_3/(m^*/m) \text{ K nm}^{-2}$ .

This result is in marked contrast with previous heat capacity studies at comparable  $^4\text{He}$  coverages on a Nuclepore substrate [19], which found  $\gamma$  to be strongly dependent on  $^3\text{He}$  coverage for  $n_3 < 1.3 \text{ nm}^{-2}$ . This was

attributed to 2D condensation, since in this case the measured  $\gamma$  is proportional to the area occupied by the 2D liquid patches, and hence  $n_3$ . However, condensation was not observed in subsequent magnetization measurements [7], also on Nuclepore. We can conclude, from that work and the present measurements, that there is no instability of the uniform 2D fluid down to 2 mK for the range of substrate and  $^4\text{He}$  film conditions studied.

The coefficient  $\gamma$  is determined from fitting the heat capacity to  $c = \gamma T + AT^2$ , and from this the effective mass is inferred [24]. In Fig. 3 we plot the coverage dependence of the reduced density of states, which for a single 2D fluid is just  $m^*/m$ . While in pure  $^3\text{He}$  films the  $^3\text{He}$  effective mass arises solely from interatomic correlations, in the present case even a single adsorbed  $^3\text{He}$  atom has an enhanced mass,  $m_H$ , which arises microscopically from its coupling to excitations of the  $^4\text{He}$  film, and which can also be visualized as arising from hydrodynamic backflow of the fluid  $^4\text{He}$  past the  $^3\text{He}$  impurity. Combining these effects one has  $m^* = m_H(1 + F_1^s/2)$ , where the density dependence arising from quasiparticle interactions is incorporated in the Landau parameter  $F_1^s$ . It is tempting to make a simple linear extrapolation of the  $m^*$  data to  $n_3 = 0$  in order to determine  $m_H$  [25]. However, according to the Fermi gas theory in 2D for  $s$ -wave repulsive interactions [26,27],  $F_1^s = 4g^2$  where  $g = 1/\ln(n_0/n_3)$  is the appropriate expansion parameter. This theory well describes the observed approximately linear dependence of  $m^*$  on  $^3\text{He}$  coverage but also features a strong variation in  $m^*$  at lower coverages ( $n_3 < 0.02n_0$ ) which should be taken into account in the determination of  $m_H^*$  in the  $n_3 \rightarrow 0$  limit; see Fig. 3 (inset). We find  $m_H = 1.40 \pm 0.03m_3$  from this procedure, in good agreement with theory [16,17], which obtains  $m_H = 1.35m_3$  for a  $^4\text{He}$  film consisting of two solid plus two fluid layers on graphite [28]. We note that the value of  $n_0$  obtained from the fit to all the points shown is  $42 \pm 7 \text{ nm}^{-2}$ . Thus  $n_3 < 0.1n_0$  over the coverage range of interest and the gas approximation should

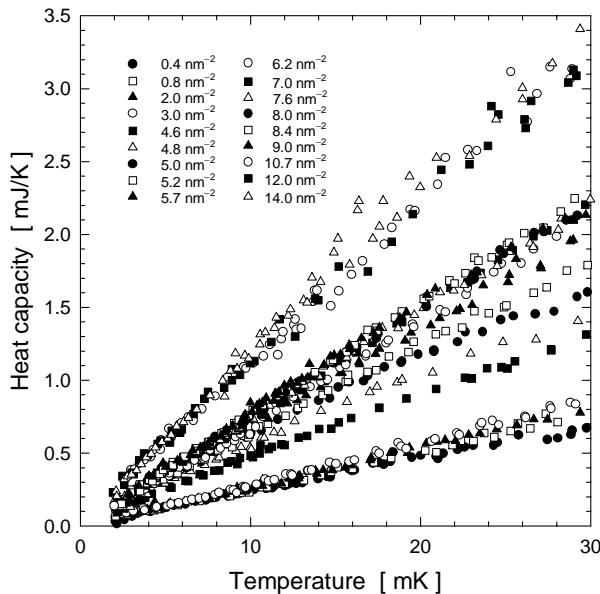


FIG. 1. Measured heat capacity as a function of temperature, for selected  $^3\text{He}$  coverages.

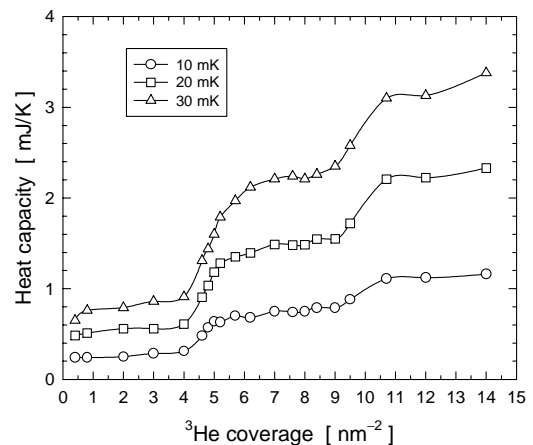


FIG. 2. Heat capacity isotherms showing steplike structure. Lines are guides to the eye.

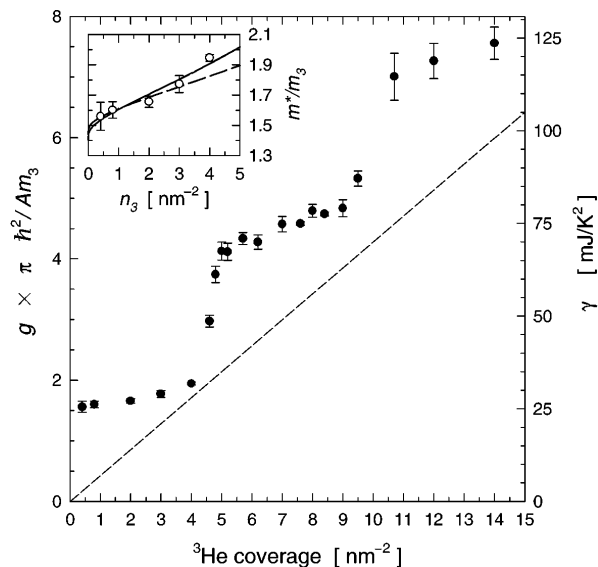


FIG. 3. Reduced density of states inferred from linear coefficient of heat capacity, as a function of  ${}^3\text{He}$  coverage. The dashed line shows the corresponding  $\gamma$  for bulk liquid at zero pressure. The inset shows surface density dependence of effective mass for  $n < n_s$  with fits to Fermi gas model. Solid line: all data; dashed line: ignoring point at  $4 \text{ nm}^{-2}$ .

remain valid [29]. As expected the hydrodynamic effective mass is comparable to that inferred for the Andreev state at the surface of bulk liquid  ${}^4\text{He}$  and smaller than that for  ${}^3\text{He}$  impurities dissolved in bulk  ${}^4\text{He}$ , for which  $m_H/m_3 = 2.2$  at zero pressure [30,31].

Presently available calculations [18] find values of  $F_1^s$  that decrease with increasing film thickness, with a magnitude comparable to that observed here. It is striking that here we find  $F_1^s = 0.7$  at  $n_3 = 4 \text{ nm}^{-2}$ , while at the same coverage in the second layer on bare graphite [22]  $F_1^s = 3.6$ . This illustrates the strong dependence of quasiparticle interactions in 2D on the surface. We plan to repeat the present experiments with a different number of  ${}^4\text{He}$  layers to further investigate the systematics of these effects.

We now turn to the steps in the heat capacity observed for  $n_3 > 4 \text{ nm}^{-2}$ . The first step is directly comparable to that seen previously in the magnetization [7], whereas the second was unresolved in that work. The first step corresponds to occupation of the first excited surface-normal state of  ${}^3\text{He}$  in the film and hence the formation of a second 2D Fermi system. With the effective mass determined experimentally here, we can determine the energy difference  $\Delta_1$  between the binding energies of the ground and first excited surface-normal states from the coverage at the step, since  $T_F = \Delta_1$ . Using the coverage of  $4.6 \text{ nm}^{-2}$  at the center of the step we find  $\Delta_1 = 1.2 \text{ K}$ . This is significantly smaller than the theoretical energy gap for the single impurity states  $\approx 2 \text{ K}$  [15–17], which is broadly consistent with values inferred from  $T_1$  measurements [8,9] at a  ${}^3\text{He}$  coverage of  $0.6 \text{ nm}^{-2}$ . This strongly indicates that  $\Delta_1$  is a decreasing function of

$n_3$ . This effect has been predicted [32] as arising from  ${}^3\text{He}$ - ${}^3\text{He}$  interactions mediated by ripplon exchange. This dominates the long range attractive interaction, and shifts the energies of the surface-normal states. For a four layer film the theory finds that  $\Delta_1$  decreases from  $\sim 3 \text{ K}$  in the single impurity limit to  $\sim 1.1 \text{ K}$  at  $n_3 = 3 \text{ nm}^{-2}$  and is reasonably constant thereafter, in good quantitative agreement with the present data.

A simple model of noninteracting fermions shows that the width of the step is consistent with thermal broadening [33]. The  ${}^3\text{He}$  coverage at the step is in reasonable agreement with that obtained from the magnetization isotherm [34].

From the magnitude of the heat capacity step, Fig. 3, we can estimate, for the first time, the hydrodynamic mass of the  ${}^3\text{He}$  quasiparticles in the first excited state. This is  $2.0 \pm 0.1 m_3$ , significantly greater than the hydrodynamic mass of the ground state. This result supports the picture of a surface-normal  ${}^3\text{He}$  state, whose wave function is located somewhat closer to the solid substrate than the ground state, having a greater overlap with that of the  ${}^4\text{He}$  film. This agrees with the interpretation of NMR measurements [8] which find that the spin lattice relaxation rate is about 50 times greater in the first excited state than in the ground state, indicative of closer proximity to the substrate. The effective mass of the first excited state is close to that calculated in the density functional theory [17], but significantly larger than that found by first-principles calculations [15]. The slope of the reduced density of states on the second plateau is approximately twice that of the first plateau. This implies an increase in the quasiparticle interactions, which may arise from coupling between two Fermi systems.

At  $9 \text{ nm}^{-2}$  we observe the onset of a second step in the heat capacity which is the signature of the formation of a further 2D continuum built on a third surface-normal state, the existence of which has been predicted theoretically [15,17]. The coverage at which this step occurs allows an estimate of the energy of this state with respect to the ground state,  $\Delta_2$ , under the assumption that  $\Delta_1$  remains constant. We find  $\Delta_2 - \Delta_1 \sim 0.4 \text{ K}$  in reasonable agreement with the single impurity energy difference calculated in density functional theory, but somewhat larger than the result of [15]. Just prior to this step the population of the first excited state is estimated to be  $\sim 20\%$ . On the basis of the available data, the hydrodynamic mass in this state is  $\sim 2 m_3$ . The fact that it was not observed in the magnetization measurements might be attributable to a combination of the relatively small value of  $\Delta_2 - \Delta_1$ , variation in binding energy due to surface heterogeneity, and the relatively high minimum temperature of that work.

In conclusion, the present study of helium mixture films on graphite reinforces their promise for studies of interactions in 2D Fermi fluids at low millikelvin temperatures, in a system of purity and simplicity, with

the possibility of readily tuning the interaction strength. Low density 2D Fermi fluids are accessible, without the intervention of 2D condensation for  $T > 2$  mK and in the absence of localization due to residual substrate heterogeneity, for testing theories of interacting fermions in the dilute gas limit and beyond. In the present work we find that the quasiparticle effective mass varies with  $^3\text{He}$  surface density in a way consistent with short range  $s$ -wave interactions. Measurements of the magnetization enhancement, when combined with  $m^*$  values, determine  $F_0^a$ . The interrelationship of the two Landau parameters should provide a key way of testing the validity of interaction models. The experimental results also confirm the importance of long range attractive ripplon mediated interactions between quasiparticles that are the analog of phonon mediated electron-electron coupling.

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