

Two-dimensional Fermi liquid in the highly correlated regime: The second layer of ^3He adsorbed on graphite

K.-D. Morhard, C. Bäuerle, J. Bossy, Yu. Bunkov, S. N. Fisher, and H. Godfrin

*Centre de Recherches sur les Très Basses Températures, Centre National de la Recherche Scientifique,
Boîte Postale 166, 38042 Grenoble Cedex 9, France*

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We present results on the magnetic susceptibility of the second atomic layer fluid of ^3He adsorbed on graphite over a large temperature and coverage range. The fluid is found to display the well characterized behavior of a Fermi liquid. The temperature dependence of the magnetic susceptibility can be described well by Dyugaev's phenomenological theory of Fermi liquids. The effective Fermi temperature has been determined as a function of coverage. We find that the susceptibility enhancement factor (with respect to the Fermi gas of the same density) reaches values larger than in bulk liquid ^3He in high-areal-density fluids. In this regime the films constitute experimental model systems for the theory of strongly correlated fermions. Using previous heat-capacity measurements performed on the same system allows a direct comparison with the predictions of the quasilocalized model and the paramagnon model.

I. INTRODUCTION

The ^3He atom is a fermion due to its nuclear spin $\frac{1}{2}$. Bulk liquid ^3He is the canonical example for Fermi liquids. Its thermal and magnetic properties have been studied extensively^{1,2} and are described well by the phenomenological theory proposed by Landau. By applying an external pressure, one can vary the density of bulk liquid ^3He in a limited range where the reduced effective mass varies between 2.8 and 5.85 and the susceptibility enhancement between 9.18 and 23.7.³ The reduced effective mass is defined as m^*/m , where m^* is the effective mass of the quasiparticles in the Landau theory, and m the mass of a ^3He atom. The susceptibility enhancement $\chi(0)/\chi_0(0)$ relates the zero-temperature susceptibility of the system $\chi(0)$ to that of the ideal Fermi gas of the same density $\chi_0(0)$.

A much larger density range can be explored in ^3He films of atomic thickness adsorbed on a solid substrate. Due to the relatively strong adsorption potential, the atomic motion is confined to two dimensions. The areal density, measured in atoms/Å², can be controlled experimentally. Contrarily to what is observed in three dimensions (3D), condensation is forbidden in the two-dimensional system.⁴ As a result, the areal density of the fluid film can be varied continuously from the very dilute noninteracting regime to the dense highly correlated regime. These new Fermi liquids have been investigated by heat-capacity^{5,6} and NMR techniques.⁷⁻⁹ At low temperatures, the heat capacity is linear in temperature, and the magnetic susceptibility is constant. These properties are common to both 2D and 3D Fermi liquids.

Motivated by previous NMR studies performed on preplated films,⁸ we have investigated the magnetic behavior of pure ^3He films. One of the advantages of pure ^3He films is that their structure is well established by neutron diffraction. In addition, heat-capacity data from which the effective mass is obtained are only available for pure ^3He films. In order to test the applicability of theoretical models to 2D ^3He films, both susceptibility and heat-capacity data for the same system are required.

II. EXPERIMENTAL DETAILS

The nuclear susceptibility of ^3He adsorbed on Grafoil, an exfoliated form of graphite, has been measured in the temperature range 3.5 to 300 mK for 12 coverages: 23.98, 26.17, 27.41, 28.63, 29.15, 29.67, 30.02, 30.37, 30.73, 31.09, 31.43, and 31.79 cc STP. The experiments were performed using a dilution refrigerator. A melting curve thermometer was used to determine the temperature with an accuracy better than 0.3% in the whole temperature range. The amount of ^3He needed to complete the commensurate phase at submonolayer coverages⁹ was 11.17 cc STP. The surface area of the sample determined by the commensurate phase coverage is 47.21 m². The Grafoil sample was contained in a plastic cell surrounded by a NMR coil which forms a resonant circuit with the capacitance of the coaxial line, with a resonance frequency of 510.5 kHz. A continuous wave radio-frequency NMR spectrometer was used to determine the nuclear susceptibility of ^3He by numerical integration of the in-phase component of the NMR signal as the external magnetic field was swept. The amplitude of the sweep was on the order of 1 mT ensuring a signal loss lower than 2%. A detailed account of the experimental techniques can be found elsewhere.^{9,10} An additional run was performed at a submonolayer coverage (16.31 cc STP) where the nuclear susceptibility is known to follow the Curie law.¹⁰ This provides a calibration of the spectrometer sensitivity for the magnetization measurements.

III. RESULTS AND DISCUSSION

For the 12 coverages investigated here, the system consists of two atomic layers of ^3He . The first layer is a high-density solid of triangular structure. Its susceptibility follows the Curie law in our temperature range.^{10,11} This solid layer acts as a new substrate for the adsorption of the second layer. The adsorption potential is considerably smoothed out by this plating compared to that of bare Grafoil.

The second layer is fluid. At very low temperatures

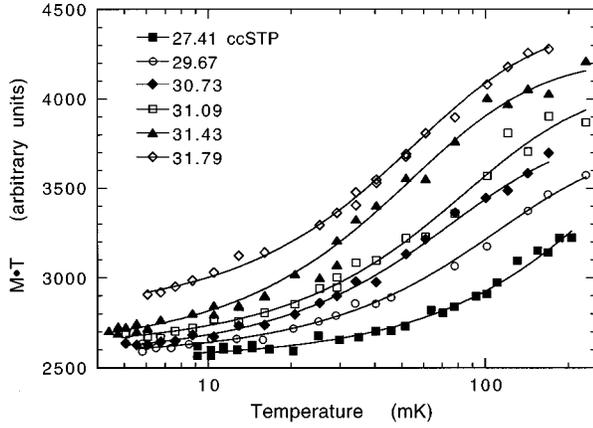


FIG. 1. Magnetization multiplied by temperature for ^3He films adsorbed on graphite at several coverages above monolayer completion. The fits are described in the text. The temperature dependence is described well by Dyugaev's theory of Fermi liquids.

($T < T_F$) the susceptibility of the fluid is relatively small and constant. For $T > T_F$ the fluid susceptibility is comparable to the solid contribution, and follows the Curie law.

This behavior is clearly seen in Fig. 1, where we show the product of the magnetization and temperature for some of the raw experimental data as a function of temperature. The first layer contribution (a horizontal line in this plot) is determined by the low-temperature data. The small increase as a function of coverage seen in the low-temperature regime is due to the first layer compression. This increase of the Curie contribution of the first layer agrees well with neutron scattering data¹² for the first layer density in the bilayer regime. For the highest coverage (31.79 cc STP), the low-temperature contribution increases substantially. This corresponds to the onset of the second layer solidification, as clearly demonstrated by heat-capacity measurements⁶ and confirmed by our NMR data at higher coverages, not shown here. The deviation of the magnetization from the Curie law is dominated by the liquid contribution. One observes that it increases with coverage, as expected from the larger number of atoms. In addition, there is a clear shift of the curves toward lower temperatures, indicating a decrease of the characteristic degeneracy temperature.

The signal of the fluid is found to be very well described by the phenomenological expression for the susceptibility of a Fermi liquid¹³

$$\chi = \frac{C_{\text{liquid}}}{\sqrt{T^2 + T_F^{**2}}} \quad (1)$$

over the whole temperature range. This expression defines the effective Fermi temperature T_F^{**} . It should be pointed out that it also provides an excellent fit of the susceptibility of bulk liquid ^3He . The expression for the modified Fermi gas used in earlier works⁸ deviates substantially from the data, especially at high densities and for temperatures somewhat larger than the effective Fermi temperature.

An important parameter for the data analysis is the areal density of the second layer liquid. Previous heat-capacity experiments⁶ provided only estimated values for the second

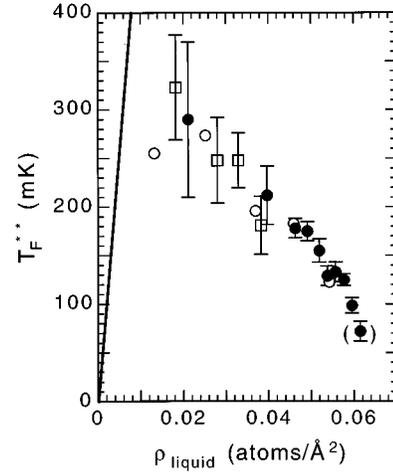


FIG. 2. Effective Fermi temperature T_F^{**} as a function of the liquid density; the solid line corresponds to the behavior of the ideal Fermi gas; filled circles: this work; open squares: ^3He on graphite (Ref. 9); open circles: ^3He on ^4He preplated graphite (Ref. 8).

layer density. It is not possible to determine the actual number of atoms in the first or in the second layer by this technique. Our NMR data at the lowest temperatures provide a direct measurement of the number of atoms in the first layer N_1 even when liquid is present in the second layer. This can be seen in Fig. 1. The number of atoms in the second layer N_2 is given with an accuracy better than 2% by $N_2 = N_{\text{tot}} - N_1$, where N_{tot} is the known total number of atoms. It would be inaccurate to calculate the density of the second layer by simply dividing the number of atoms in the second layer by the submonolayer commensurate phase area given above. In fact, the effective area of the substrate increases slightly with coverage, as shown by neutron scattering¹² and NMR experiments.¹⁰

We therefore determine the second layer density n_2 as $n_2 = n_1 N_2 / N_1$, using the first layer density n_1 measured at second layer coverages by neutron diffraction.¹² With this procedure we obtain values for the second layer density with an accuracy of 3%.

The dependence of T_F^{**} on the areal density of the second layer liquid is shown in Fig. 2. The effective Fermi temperature decreases as a function of coverage in the density range from 0.02 to 0.06 atoms/ \AA^2 . Its magnitude is practically identical to that observed in the $^3\text{He}/^4\text{He}$ preplated system⁸ and in submonolayer ^3He on Grafoil.⁹ This clearly shows that the strength of the adsorption potential (different for the three systems) does not strongly influence the magnetic properties of the ^3He films. This is in contrast with the strong effect of the potential observed on the quantum exchange in solid films.^{10,14} Therefore a single parameter T_F^{**} , dependent on the density, characterizes the magnetic susceptibility of fluid ^3He films of atomic thickness. It would be interesting to investigate the magnetic properties of the fluid in the third layer where the adsorption potential is even weaker.

We observe a rapid decrease of the effective Fermi temperature for areal densities in the range from 0.05 to 0.06 atoms/ \AA^2 . At higher densities one observes a gradual solidification of the second layer. Although it is not within

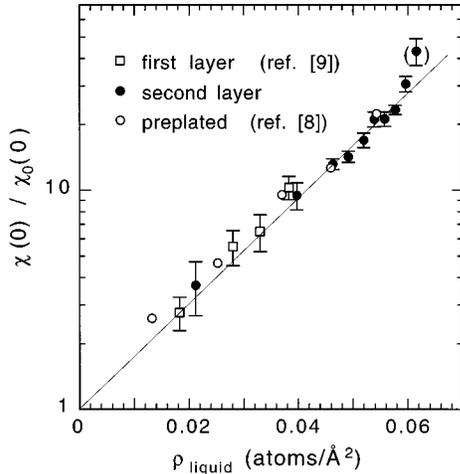


FIG. 3. Susceptibility enhancement as a function of the liquid density; filled circles: this work; open squares: ^3He on graphite (Ref. 9); open circles: ^3He on ^4He preplated graphite (Ref. 8).

the scope of the present article to discuss this process, one should mention two possible effects. First, solidification may be induced by heterogeneity. Regions of higher adsorption potential can localize some fraction of the adsorbed fluid at relatively low coverages. Previous measurements of the first layer of ^3He adsorbed on Grafoil⁹ showed that up to 4% of the atoms are localized. For the second layer coverages investigated here we find that this effect is negligible (less than 2%) due to the smoothing of the adsorption potential by the first layer plating. A second effect, the thermal growth of the solid, requires some care in the analysis of the data obtained for densities close to solidification. For coverages below 31.43 cc STP the areal density of the fluid is sufficiently low to exclude this type of solidification. The presence of solid at low temperatures is clearly seen in the data at a coverage of 31.79 cc STP. These are consistent with an effective Fermi temperature similar to that observed before the onset of solidification, as expected for a fluid-solid coexistence regime. The point shown in parentheses in Fig. 2 corresponds to data at a coverage 31.43 cc STP where there is clearly no solid in the second layer at low temperatures, as seen in Fig. 1. However, since this coverage is close to the onset of solidification, we cannot exclude the possibility that some solid is formed as the temperature is increased. This effect would be similar to that observed in bulk liquid ^3He due to the negative slope of the melting curve. (See also Fig. 3.)

It should be pointed out that the results on preplated films shown in Figs. 5 and 6 seem to agree well with the data on the pure ^3He films investigated here. However, the effective mass values used in the analysis of the preplated films data are taken for densities that agree neither with those estimated by Greywall^{6,15} nor with our corrected values, as seen in Fig. 4. This is not a severe problem for the analysis of the data for low-density liquids. At high densities, however, the strong density dependence of the effective mass makes the agreement only qualitative since substantial error bars should be added to the preplated films data in this range.

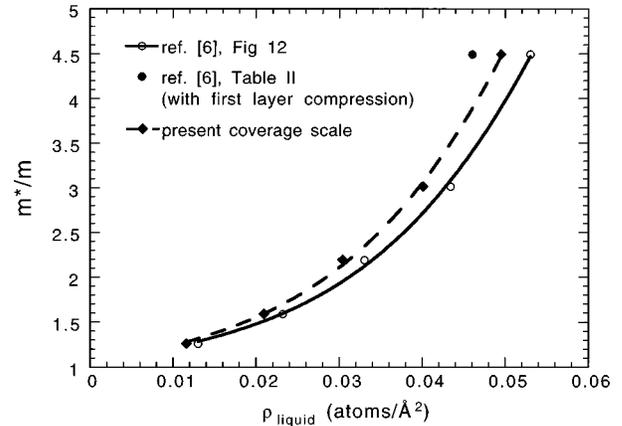


FIG. 4. Reduced effective mass as a function of the second layer density. Open circles: data from Ref. 6; filled circle: represents the estimated coverage correction due to the compression of the first layer as given in Table II in Ref. 6; filled diamonds: same heat-capacity data as a function of the areal density of the second layer determined in the present work.

IV. CONCLUSIONS

The second layer fluid of ^3He adsorbed on graphite is a particularly interesting two-dimensional system where the effect of heterogeneity is considerably attenuated by the plating effect of the first solid layer.

The magnetic susceptibility of this system has been investigated by NMR measurements. The Curie contribution for the first layer is found to increase slightly with coverage in the bilayer regime in agreement with the first layer compression measured directly by neutron diffraction.

The temperature dependence of the magnetic susceptibility of the second layer fluid is described well for a wide

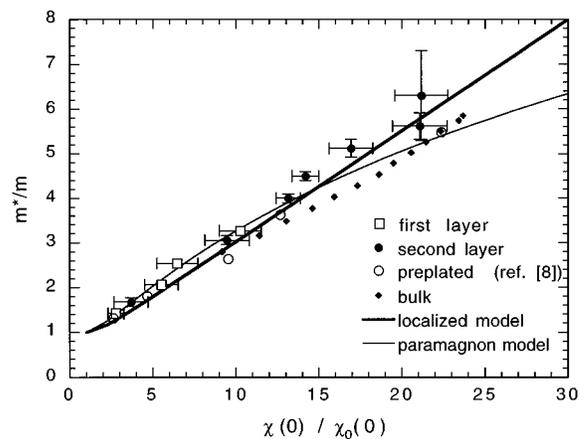


FIG. 5. The reduced effective mass as a function of the susceptibility enhancement; filled circles: this work; open squares: ^3He on graphite (Ref. 9); open circles: ^3He on ^4He preplated graphite (Ref. 8); filled diamonds: bulk ^3He (Ref. 3). The thick line corresponds to the quasilocated model (Ref. 17), and the thin line to the paramagnon model (Ref. 16). Our susceptibility enhancement factors larger than 22 cannot be represented in this plot due to the lack of heat-capacity data. Note that for these points very different effective masses are predicted by the different models.

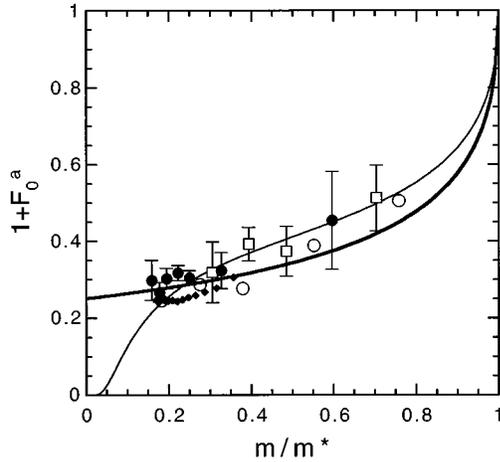


FIG. 6. The Landau parameter $1 + F_0^a$ as a function of the inverse reduced effective mass; filled circles: this work; open squares: ^3He on graphite (Ref. 9); open circles: ^3He on ^4He preplated graphite (Ref. 8); filled diamonds: bulk ^3He (Ref. 3). The thick line corresponds to the quasilocalized model (Ref. 17), and the thin line to the paramagnon model (Ref. 16).

range of densities by the expression proposed by Dyugaev with a single parameter T_F^{**} , the effective Fermi temperature. On the other hand, the modified Fermi gas expression used in earlier works does not fit our data adequately in the whole temperature range.

We observe that the effective Fermi temperature decreases with increasing coverage for the entire coverage range investigated here. A comparison with our previous results obtained on submonolayer films shows that the magnitude of T_F^{**} for a given density is rather insensitive to the strength of the substrate potential.

We find that the susceptibility enhancement with respect to the Fermi gas of the same density displays an exponential dependence as a function of the areal density. At the highest densities we observe that the susceptibility enhancement is stronger than in the bulk liquid. Values as high as 30 have been determined in this regime, not investigated up to now. Even values as high as 40 are possible, but the uncertainty of the structure of the films does not allow us to analyze the susceptibility data unambiguously in this regime as explained above.

Our results at moderate densities agree well with previous ones obtained on samples preplated by ^4He . However, the contention that the quasilocalized model is strongly supported by these data is not valid; we find that both the quasilocalized and the paramagnon models provide a reasonable description of the data in this coverage range.

These models, however, make substantially different predictions for higher-density films where our measurements reveal the existence of susceptibility enhancement factors larger than 30. The predictions of the quasilocalized model and of the paramagnon model for the effective mass in this coverage range should be tested experimentally by further heat-capacity measurements. These measurements, combined with the ones reported here, should then establish whether the physics of fluid ^3He is dominated by quasisolid or quasi-ferromagnetic effects. This system where the interaction parameter can be varied continuously from the noninteracting gas to the strongly correlated regime can also be used to test other theoretical models of correlated fermions.

ACKNOWLEDGMENTS

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¹J. Wilks, *Liquid and Solid Helium* (Clarendon, Oxford, 1967).
²D. Pines and P. Nozières, *The Theory of Quantum Liquids* (Benjamin, New York, 1966).
³Tabulated values in D. Vollhardt and P. Wölfle, *The Superfluid Phases of ^3He* (Taylor and Francis, London, 1990), p. 31.
⁴M. D. Miller and L. H. Nosanow, *J. Low Temp. Phys.* **32**, 145 (1978).
⁵M. Bretz, J. G. Dash, D. C. Hickernell, E. O. McLean, and O. E. Vilches, *Phys. Rev. A* **8**, 1589 (1973).
⁶D. S. Greywall, *Phys. Rev. B* **41**, 1842 (1990).
⁷J. Saunders, C. P. Lusher, and B. P. Cowan, in *Excitations in Two-Dimensional and Three-Dimensional Quantum Fluids*, edited by A. G. F. Wyatt and H. Lauter (Plenum, New York, 1991).
⁸C. P. Lusher, B. P. Cowan, and J. Saunders, *Phys. Rev. Lett.* **67**, 2497 (1991).
⁹K.-D. Morhard, J. Bossy, and H. Godfrin, *Phys. Rev. B* **51**, 446 (1995).
¹⁰R. E. Rapp and H. Godfrin, *Phys. Rev. B* **47**, 12 004 (1993).

¹¹P. Schiffer, M. T. O'Keefe, D. D. Osheroff, and H. Fukuyama, *Phys. Rev. Lett.* **71**, 1403 (1993); *J. Low Temp. Phys.* **94**, 489 (1994).
¹²H. J. Lauter, H. Godfrin, V. L. P. Frank, and H. P. Schildberg, *Physica B* **165&166**, 597 (1980).
¹³A. M. Dyugaev, *Sov. Sci. Rev. A Phys.* **14**, 1 (1990).
¹⁴H. Franco, R. E. Rapp, and H. Godfrin, *Phys. Rev. Lett.* **57**, 1161 (1986).
¹⁵Large corrections to the second layer density have been estimated by Greywall (Ref. 6). The values given in Table II of Ref. 6 (where the effect of the first layer compression is taken into account) differ substantially from those of Fig. 12 used to analyze earlier NMR work (Ref. 8).
¹⁶M. T. Béal-Monod and A. Theumann, in *Ordering in Two Dimensions*, edited by S. K. Sinha (Elsevier North Holland, New York, 1980).
¹⁷D. Vollhardt, *Rev. Mod. Phys.* **56**, 99 (1984).