RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 40, NUMBER 10 1 OCTOBER 1989

Density-functional theory of thin films of self-bound fermions

R. A. Guyer, K. R. McCall, and D. T. Sprague
Laboratory for Low Temperature Physics, Department of Physics and Astronomy, University of Massachusetts, Amherst, Massachusetts 01003

(Received 5 July 1989)

Thin films of self-bound fermions, fermion oceans, show striking evidence of the evolution of the states available to the particles; the susceptibility and specific heat are an oscillatory function of the film thickness. A density-functional theory of the structure and thermodynamics of these films is developed.

Two-dimensional systems of fermions, fermion films, fall into two broad categories: the "atmospheres" that are not self-bound (gas) and held in two-dimensional conformation at a density determined by an external field and the "oceans" that are self-bound (liquid) and achieve approximately uniform density as a consequence of self-interaction. The typical fermion atmosphere is a charged system, e.g., heterostructures or electrons on ^{4}He (Ref. 1). The typical fermion ocean is uncharged, e.g., ^{3}He.

It is the latter system that we are interested in; ^{3}He films have been the subject of a wide range of investigations.[1] Both the thermodynamic and transport properties of these films have been measured in a variety of circumstances, e.g., the specific heat of ^{3}He on Grafoil[2] (on ^{4}He),[3] the surface sound mode of ^{3}He,[4] the third sound mode of ^{3}He-^{4}He mixture films,[5,6] the magnetic properties of ^{3}He films on Grafoil[7-9] (on ^{4}He),[10] etc. In this paper we sketch a density-functional theory of these self-bound films that provides a good qualitative understanding of the existing experimental data and lets us predict the behavior of a wide variety of thermodynamic and transport properties.

We consider a ^{3}He film made up of N ^{3}He atoms on the surface of a ^{4}He film[11] of area A. For N of order or greater than a "monolayer" ($N \gtrsim A/a_3^2 \equiv N_0$, where a_3 is the interparticle spacing in low-pressure bulk ^{3}He), the ^{3}He film is self-bound. Take the single-particle states available to the ^{3}He atoms to be those of a box of the atoms' own making. That is, we take the N atoms of the film to be confined to a region of space of size Ad and assign them, in accordance with the Pauli principle, to the single-particle states

$$\varphi_{\mathbf{k},m}(\boldsymbol{\rho},z) \propto \sin\frac{m\pi z}{d}\exp(i\mathbf{k}\cdot\boldsymbol{\rho}) ,$$

having energy

$$\varepsilon_{\mathbf{n},m} = \varepsilon_0\left[\frac{n^2}{N_0}+\frac{m^2}{(d^*)^2}\right] , \tag{1}$$

where

$$\varepsilon_0 = \frac{\hbar^2\pi^2}{2m_3a_3^2},\quad N_0 = A/(a_3^2),\quad \mathbf{k} = \pi\mathbf{n}/\sqrt{A},\quad d^* = d/a_3 .$$

In addition to the kinetic energy associated with occupying the single-particle states, the atoms have an energy of interaction that causes a suitably large number of them to attempt to achieve bulk ^{3}He density. For this interaction energy, we take the density-functional approximation[12,13]

$$V(d,N) = \int v_0\left[-2\left(\frac{n(z)^2}{N_0d}\right)+\left(\frac{n(z)^3}{N_0^2d^2}\right)\right]dz , \tag{2}$$

where

$$n(z) = \sum_m N_m\phi_m(z)^2 ,\quad N_m = \sum_{\mathbf{n}} n(\varepsilon_{\mathbf{n},m},\varepsilon_F) ;$$

$n(\varepsilon_{\mathbf{n},m},\varepsilon_F)$ is the occupation probability of the state $(\mathbf{n},m)$ and ε_F is the Fermi energy. Thus, the free energy of the system at finite temperature is taken to be

$$F(T,N;d) = \sum_\alpha \varepsilon_\alpha n_\alpha - TS + V(N,d) , \tag{3}$$

where S is the entropy,

$$S = -k_B[n_\alpha \ln n_\alpha + (1-n_\alpha)\ln(1-n_\alpha)] , \tag{4}$$

$\alpha = (\mathbf{n},m)$, ε_α is given by Eq. (1), and $n_\alpha = n(\varepsilon_\alpha,\varepsilon_F)$. At fixed (N,d), $n_\alpha = \{\exp[\beta(\varepsilon_\alpha-\varepsilon_F)+1]\}^{-1}$ with the requirement

$$N = \sum_\alpha n_\alpha , \tag{5}$$

fixing ε_F. To place the film in a substrate potential, $V(z)$, we would add $\int dz\, V(z)n(z)$ to Eq. (3). However, in this paper, for simplicity, we drop this term[14] and work with a model that is valid for $N/N_0 \gtrsim 1.0$.

We put $T=0$ and examine $E(N;d)$. At fixed N, we minimize $E(N;d)$ with respect to d; i.e., we find the best size (thickness) for a film of N particles. For $N \gg N_0$, the sum on m goes over to an integral, $n(z)$ is essentially constant, and we find $d \propto N/N_0$. We use the result in this limit to choose v_0 so that $E/N \sim -3$ K, the binding energy per particle of bulk ^{3}He. Turning to thin films, $N \gtrsim N_0$, we find the single-particle energies, Fermi energy, film thickness, and magnetization shown in Fig. 1.[15] These results are understood with the help of Fig. 2 in which we show the $\mathbf{k}$ space, $(\mathbf{n},m)$ space, for the states available to the particles. Because we are dealing with thin films, $1 \lesssim d^* \lesssim 10 \ll \sqrt{N_0} \simeq 10^8$, the m levels are separated by energies comparable to both the Fermi energy and the interaction energy associated with the structure of the film. Occupied energy levels lie in a small

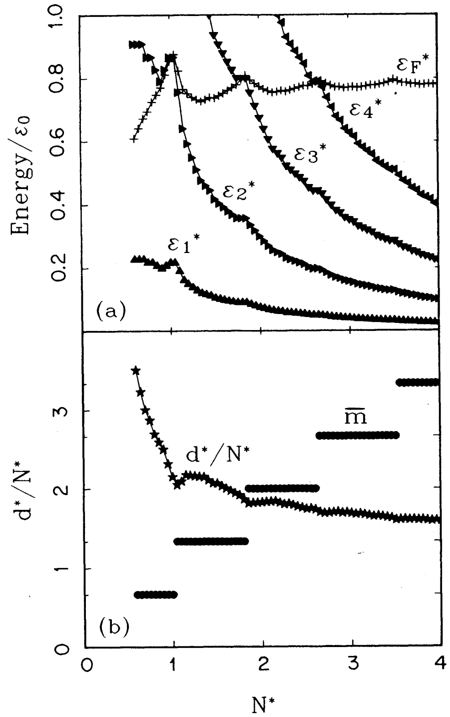

FIG. 1. $T=0$ behavior of fermion film. (a) Dimensionless Fermi energy ε_F^* and energies of the four lowest-lying states in the film (crosses and triangles, respectively) vs N/N_0; no substrate potential is applied. The $m=2$ disk enters the Fermi sphere at the point at which ε_F^* crosses the ε_2^* curve and the magnetization steps up by one unit. This event is not seen dramatically in the behavior of the total energy or film thickness. (b) Dimensionless film thickness $d^*=d/a_3$ vs N/N_0. For $N/N_0>1.0$, $d^* \propto N/N_0$, a result that can be found from asymptotic analysis of Eq. (3). There is modest evidence in d^* for the evolution of occupation of the states in the film. Shown also is $\bar m$, the largest m state within the Fermi sphere, vs N/N_0.

number of disks, $1 \leq m \leq \bar m$. The disk has a radius of $\bar n(m) = [N_0(\varepsilon_F^* - m^2/d^{*2})]^{1/2}$. As N is increased, the film adjusts its thickness and thereby the single-particle energies, the size of the disks, and the density, to achieve minimum energy. The single-particle energies and ε_F are shown as a function of $N^* = N/N_0$ in Fig. 1(a). With an increase in N^*, the disks in $(\mathbf{n},m)$ space shrink toward the origin and more disks fit within the Fermi sphere. Properties of the film that are sensitive to the occupation of the disks will show direct evidence of these events in $(\mathbf{n},m)$ space. The magnetization and low-temperature heat capacity (see later) act approximately as counters of the number of disks within the Fermi sphere. We see this in Fig. 1(b) where we show d^*/N^* and $\bar m$ vs N^* for $T=0$. Because $\mu H \ll \varepsilon_{m+1} - \varepsilon_m$, the particles in the film respond to an external magnetic field as $\bar m$ independent systems of fermions, each two dimensional. Each such system of fermions, e.g., the mth, has magnetization M_m proportional to $N_m \mu^2 H/\varepsilon_F^m$, where $N_m \propto \bar n(m)^2$ is the number of particles in the mth disk and ε_F^m is the "Fermi energy of the particles in the mth disk." Since $\varepsilon_F^m \propto (\hbar^2/m_3)(N_m/A)$, M_m is constant, independent of N.

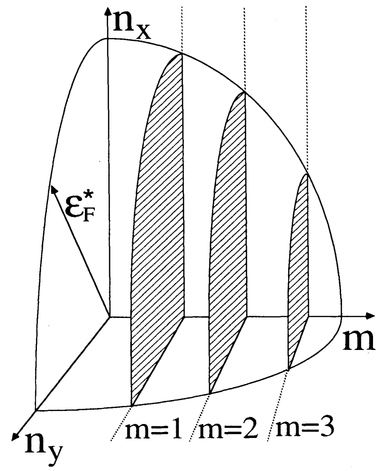

FIG. 2. **k** space. The states available to the particles are closely spaced in Δn since $N_0 \simeq 10^{15}$ and widely spaced in Δm since $d^* \simeq 1$. As a consequence, the particles occupy states in disks of radius $\bar n(m) = \{N_0[\varepsilon_F^* - m^2/(d^*)^2]\}^{1/2}$. As more particles are added to the system the disks move toward the origin and their spacing decreases. When a new disk enters the Fermi sphere, which changes only modestly in radius with N/N_0, the magnetization and heat capacity step upward.

Thus the total magnetization,

$$M = \sum_{m=1}^{\bar m} M_m \propto \frac{m_3 A}{\hbar^2} \sum_{m=1}^{\bar m} 1 \propto \bar m ,$$

steps up by one unit as each disk enters the Fermi sphere.

Let us turn to the discussion of the $T \neq 0$, low-temperature thermodynamic properties of the film. Upon minimizing $F(T,N;d)$ with respect to d at fixed N and T, we find that, for $T^* \ll 1$, the evolution of the film is not importantly effected by finite T, i.e., $d(T,N) \simeq d(0,N)$. Thus, to describe the $T \neq 0$ thermodynamics, we use a truncated form of the free energy: the first two terms of Eq. (3) with the constraint $d = d(0,N)$.

Straightforward but lengthy calculations lead to formulas for the low-temperature magnetization, heat capacity, pressure, and surface sound velocity. We show the result of numerical implementation of the formulas for the magnetization and heat capacity C in Fig. 3 for two values of T^*. Like the magnetization, at the lowest temperatures, C shows the step structure that follows from $C_m \propto N_m k_B T/\varepsilon_F^m$. Unlike the magnetization, C is not simply additive among the disks, $C \neq \sum C_m$, so that as T^* is raised, there is a more marked departure from the simple disk counting behavior that characterizes the behavior of the magnetization.

Two recent thermodynamic measurements lend support to the picture we have developed. Magnetization measurements (Higley, Sprague, and Hallock[16]) of ^{3}He atop two layers of ^{4}He on a Nuclepore substrate show a step structure similar to what we have described above. In the experiment, this structure washes out long before it does in our model. Part of this washing out may be due to inhomogeneity in the Nuclepore substrate. A more important part may have its source in interaction effects among particles in the various disks that are not accounted for in our model. Greywall and Busch[17] have studied the

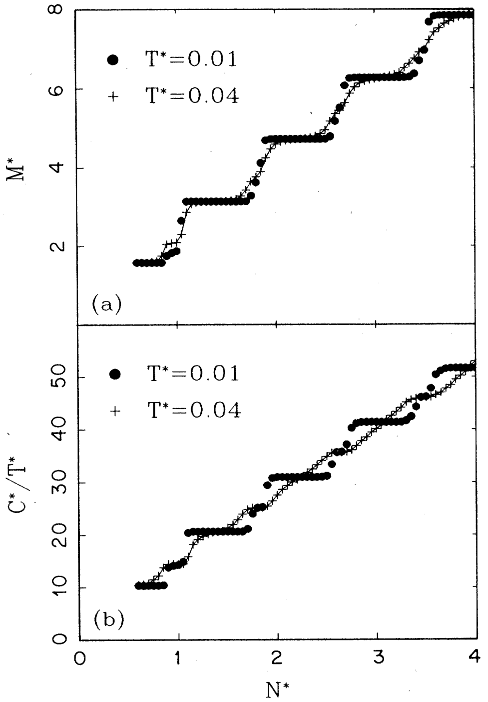

FIG. 3. Thermodynamic quantities vs N/N_0 at $T\neq 0$. Dimensionless magnetization and heat capacity are plotted vs N/N_0 for two values of dimensionless temperature. The steplike behavior of M^* and C^*/T^* would appear as oscillatory behavior in the susceptibility and in the specific heat.

specific heat of ^{3}He on Grafoil at very low temperatures, 2.0 mK$\lesssim T \lesssim$300 mK, for ^{3}He films with coverages from one to five layers. The data at 200 mK for $N/N_0>2$ "layers", e.g., ^{3}He atop two solid layers of ^{3}He on Grafoil, looks similar to the $T^*=0.04$ curve in Fig. 3. This similarity of appearance is encouraging. However, we caution that the ^{3}He system on Grafoil has a complicated phase diagram[9] that must be understood before it is possible to make an unambiguous assignment as to the source of a specific-heat feature. In addition, a third set of measurement, the specific-heat measurements of Bhattacharyya, Dipirro, and Gasparini[3] shows vestiges of the behavior we are describing.

Above, we have described a simple density-functional model of ^{3}He films and the results of calculations of thermodynamic properties within this model. What emerges is a picture in which the thermodynamic properties of these films arise from excitation of the particles in the occupied m levels. It is natural to suggest that a correct theory of the thermodynamics of thin films is a Fermi-liquid theory of interacting systems of quasiparticle excitations, e.g., at $N/N_0 \simeq 3$, the $m=1,2$, and 3 systems of quasiparticles are present in the film. The formal structure of such a theory can be built up from the work of Havens-Sacco and Widom.[18] It necessarily involves a complicated set of Fermi-liquid parameters: interactions between particles in the same state, involving $F^{m,m}$, and interactions between particles in different m states, involving $F^{m,m'}$. Some of the quantitative properties of this description can be provided by the model we have introduced above, with modifications designed to more accurately describe real films.[14] Ultimately, one would want to "found" this picture in an appropriate microscopic theory. Important progress in developing a microscopic description of these systems, for $N/N_0 \ll 1$ and only the $m=1$ disk occupied, has been made by Krotschek, Saarela, and Epstein.[19]

Now let us turn to a discussion of the transport properties of the fermion films. This subject is made complicated by the absence of experiments that direct our choice among many alternatives. For a ^{3}He film which resides on ^{4}He (^{3}He), which, in turn, resides on a Nuclepore (Grafoil) substrate, ^{3}He atoms in the film interact (a) with other ^{3}He atoms in the film, (b) with ^{3}He atoms in the ^{4}He (^{3}He) layers, (c) with the excitations that are supported by these layers, and (d) with fluctuations in the substrate potential. Generally, it is believed that the ^{3}He system is "clamped" to the substrate. The meaning of this phrase is that the ^{3}He film loses momentum on a time scale much shorter than that at which it is being probed, e.g., a typical third sound experiment on a mixture film is at 10^3 Hz. This clamping may be direct, (d), or through coupling to the ripplons, (c), which in turn are "strongly" clamped. Thus, in experiments on modes of a mixture film, the compressional mode of the ^{3}He is not observed;[4,5] the ^{3}He makes itself known only through a modification of the Q of the third sound mode.

We have constructed a theory of the involvement of ^{3}He in the determination of Q of the third sound mode that employs our theory of the structure of the film. Q vs N found from this theory oscillates as a function of N/N_0 in striking agreement with the experiment of Ellis and Hallock;[5] the peaks in Q occur at the steps in M. This theory leads to Q which is sensitive only to the heat capacity and N, independent of the ^{3}He dynamics, because of the clamping. The agreement between theory and experiment at most confirms our model for the thermodynamic properties of the film; it has nothing to say about ^{3}He dynamics.

The dynamics of thin ^{3}He films have been probed in the nuclear-magnetic-resonance experiments of Valles *et al.*[20] Unfortunately, a variety of spurious effects have made a comprehensive understanding of T_1,T_2, and diffusion data intractable. Theoretical progress on aspects of the problem at hand has been made by Saslow and Kumar,[21] Tesanovic, Jaric, and Maekawa, and Fishman and Calecki.[22]

We have introduced a density-functional model of thin fermion films that focuses attention on the self-consistent energy levels occupied by the particles in the film. This model attributes the important features in the thermodynamic properties of the film to excitations of a small number of two-dimensional systems of particles. We believe this model provides a useful starting point from which to develop appropriate Fermi-liquid and microscopic theories.

The work of K.R.M. was supported by Schlumberger-Doll Research, Ridgefield, CT.; that of D.T.S. was supported by National Science Foundation Grants No. DMR 85-17939 and No. 88-20517.

[1] T. Ando, A. B. Fowler, and F. Stern, Rev. Mod. Phys. **54**, 437 (1982).

[2] J. G. Dash, *Films on Solid Surfaces* (Academic, New York, 1975).

[3] B. K. Bhattacharyya, M. J. Dipirro, and F. M. Gasparini, Phys. Rev. B **30**, 5029 (1984).

[4] D. O. Edwards, S. Y. Shen, J. R. Eckardt, P. P. Faturos, and F. M. Gasparini, Phys. Rev. B **12**, 892 (1975); Phys. Rev. Lett. **32**, 706 (1974).

[5] F. M. Ellis and R. B. Hallock, Phys. Rev. B **29**, 497 (1984).

[6] J. P. Laheurte, J. C. Nornay, and J. P. Romagnan, J. Phys. (Paris) Lett. **42**, L197 (1981).

[7] M. G. Richards, in *Phase Transitions in Surface Films,* edited by J. G. Dash and J. Ruvalds (Plenum, New York, 1980), p. 165.

[8] L. J. Friedman, A. L. Thomson, C. M. Gould, H. M. Bozler, P. B. Weichmann, and M. C. Cross, Phys. Rev. Lett. **62**, 1635 (1989).

[9] H. Godfrin, Can. J. Phys. **65**, 1430 (1987).

[10] J. M. Valles, Jr., R. H. Higley, R. B. Johnson, and R. B. Hallock, Phys. Rev. Lett. **60**, 428 (1988).

[11] A. F. Andreev, Zh. Eksp. Teor. Fiz. **50**, 1415 (1966) [Sov. Phys. JETP **23**, 939 (1966)].

[12] R. A. Guyer, J. Low Temp. Phys. **64**, 49 (1986).

[13] G. Ji and M. Wortis, Phys. Rev. B **34**, 7704 (1986).

[14] For example, we could choose $V(z)$ so that the single-particle energies go over to the single-particle energies appropriate to a dilute film, e.g., B. K. Bhattacharyya, Ph.D. thesis, University of Buffalo, 1982. See also the work of F. M. Gasparini and collaborators on the dilute film problem, e.g., Ref. 3. We find that the qualitative results being reported are unchanged by this modification. In a longer paper we plan to describe these calculations and other elaborations of the simplest model.

[15] We use $*$ to denote dimensionless measures of energy, length, and number; energies are scaled by $\varepsilon_0 = \pi^2\hbar^2/2m_3a_3^2$, $1/a_3^3$ is the bulk density, d is scaled by a_3, N is scaled by N_0, $N_0 = A^2/a_3^2$; $T^* = k_BT/\varepsilon_0, \varepsilon_F^* = \varepsilon_F/\varepsilon_0$, etc.

[16] R. H. Higley, D. T. Sprague, and R. B. Hallock, Bull. Am. Phys. Soc. **34**, 679 (1989); and (unpublished).

[17] D. S. Greywall and P. A. Busch, Phys. Rev. Lett. **62**, 1868 (1989); and (unpublished).

[18] S. M. Havens-Sacco and A. Widom, J. Low Temp. Phys. **40**, 357 (1980).

[19] E. Krotscheck, M. Saarela, and J. L. Epstein, Phys. Rev. Lett. **61**, 1728 (1988).

[20] J. M. Valles, Jr., R. H. Higley, B. R. Johnson, and R. B. Hallock, Can. J. Phys. **65**, 1564 (1987); J. M. Valles, Jr., Ph.D. thesis, University of Massachusetts, 1987.

[21] W. M. Saslow and A. A. Kumar, Phys. Rev. B **33**, 305 (1986).

[22] Z. Tesanovic, M. V. Jaric, and S. Maekawa, Phys. Rev. Lett. **62**, 2760 (1986); G. Fishman and D. Calecki, *ibid.* **62**, 1302 (1989).