^{3}He Films and the Ruderman-Kittel-Kasuya-Yosida Interaction

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^{3}He films on Grafoil at areal density $n \gtrsim 0.25$ atom/Å^2 have unusual magnetic properties. This system is modeled as a two-dimensional solid in interaction with a two-dimensional Fermi liquid. A calculation of the magnetic behavior of this model leads to a Ruderman-Kittel-Kasuya-Yosida exchange interaction that has several features in reasonable accord with experiment.

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Several recent thermodynamic measurements[1-5] have focused attention on the neutral, two-dimensional Fermi systems that reside in thin ^{3}He-^{4}He films.

For example, (1) the specific-heat measurement of Greywall,[4] at $d_4=0$ in Fig. 1, has elaborated the already rich phase diagram of thin ^{3}He films on Grafoil.[6] Of particular interest is the second layer. This two-dimensional Fermi system[7] forms as a dilute gas at total coverage, n, equal to 0.110^+ atom/Å^2. For $0.110 < n < 0.182$ almost all particles added to the film[7] go to increase the density in this layer and to drive a fluid-solid phase transition within the layer at $n=0.168$ ($n_2=0.055$). For $n > 0.182$ almost all particles added to the film go into a liquid layer that lies atop the second (now solid) layer. Attending the increase in the number of particles in the film at $n > 0.182$ is a modest evolution in the density of the second solid layer (less than 15%) that drives a sequence of solid phase transitions, RS2 → RS2′ → IS2 (see the inset of Fig. 1). The second-layer solid system that exists at $n \gtrsim 0.18$ has striking specific-heat and magnetization features.[1-4] (2) The magnetization measurements of Higley, Sprague, and Hallock,[5] at $d_4=2.2$ in Fig. 1, are on a ^{3}He liquid layer that sits atop approximately two layers of ^{4}He on a Nuclepore substrate.[8] As the number of ^{3}He particles in the liquid layer is increased, their magnetization undergoes a sequence of quantized steps at $n=0.055$, 0.100,.... Guyer, McCall, and Sprague[9] have explained the qualitative behavior of these data, i.e., the magnetization steps, in terms of evolution of the states available to fermions in an adjustable box, the box size being driven by n.[10]

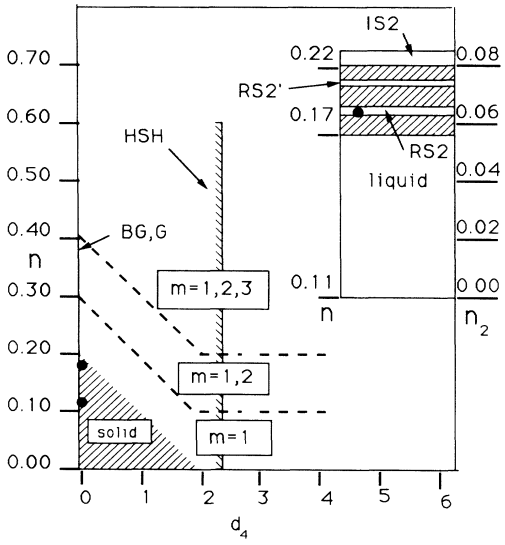

FIG. 1. (d_4,n) phase diagram. ^{3}He-^{4}He films are studied for varying thicknesses of ^{3}He (n=atom/Å^2) on ^{4}He films of varying thickness d_4 on various substrates (Ref. 7). The experiments of Greywall and Busch and of Greywall are on Grafoil at $d_4=0$; that of Higley, Sprague, and Hallock is on Nuclepore at $d_4=2.2$. The energy level structure within the liquid film at $d_4=2.2$ on Nuclepore is indicated ($m=1,\ldots$); a plausible interpolation of this structure, without regard to substrate dependence, is shown by a dashed line. The coverages of second- and third-layer promotion at $d_4=0$ on Grafoil are shown as solid circles. Inset: detail of the second-layer phase diagram for ^{3}He on Grafoil at $T=0$, n_2 vs n. Where these schematic phase diagrams are quantitative the numbers are appropriate to Grafoil and are from Greywall.

In this paper we introduce a model which unifies the description of features of the systems studied by Greywall and by Higley, Sprague, and Hallock. We argue that the ^{3}He liquid layer atop the second solid layer studied by Greywall is much the same as the ^{3}He liquid layer studied by Higley, Sprague, and Hallock. It is a system of fermions in an adjustable box. Using this model we find that the magnetic behavior of the second solid layer, at $n \gtrsim 0.25$, may be understood in terms of a Ruderman-Kittel-Kasuya-Yosida exchange interaction[11] between particles in this layer that is mediated by the liquid layer. This exchange interaction is very sensitive to the behavior of particles in the liquid layer.

We model the system with a two-dimensional hexagonal solid layer[7] of area A on which ^{3}He particles, having wave function

$$\psi_R(\mathbf{x}) = \psi_R(\boldsymbol{\rho})\psi_\perp(z), \tag{1}$$

are localized near lattice sites $\mathbf{R}_1,\ldots,\mathbf{R}_N$ (see Fig. 2). Adjacent to this layer is a liquid layer of thickness d in which ^{3}He particles move parallel to the solid layer in plane-wave states, $\psi_q(\boldsymbol{\rho}) = (1/\sqrt{A})e^{i\mathbf{q}\cdot\boldsymbol{\rho}}$. These particles

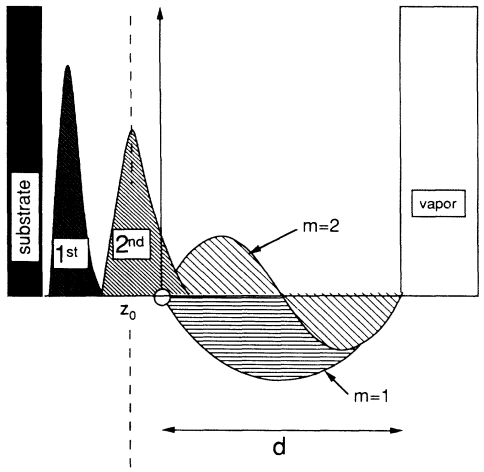

FIG. 2. Wave functions. At $n > 0.25$ atom/Å^2 the second solid layer ceases to evolve; the z part of the wave function of a particle in this layer, $\psi_\perp(z)$, sticks out into the liquid layer. In the liquid layer the particles are described by simple wave functions that evolve as n increases. The overlap integral between $\psi_\perp$ and the ψ_m leads to the n dependence of the RKKY coupling.

have perpendicular energy states, the m states, corresponding to

$$\psi_m(z) = (2/d)^{1/2}\sin(m\pi z/d)\,, \tag{2}$$

where d is a functional of n_L; $n_L = n - n_1 - n_2$, $n_1 = 0.114$, $n_2 = 0.082$. The energies of the m states, ϵ_m, and their occupation numbers, N_m, are functions of the liquid-layer thickness.[7] For small n the states occupied by the particles in the liquid layer lie on a sequence of disks, each labeled by m, of radii $q_m = (q_F^2 - \pi m^2/d^2)^{1/2}$, where q_F is the Fermi wave vector of disk m (see Fig. 2 of Ref. 9). When there are few particles in the liquid layer all of them are in the $m=1$ disk, $N_1 = n_L$. As n_L increases, the energies of the m states, $\epsilon_m \propto m^2/d^2$, decrease and at $n_L \simeq 0.07$ the Fermi energy crosses from below to above ϵ_2, the $m=2$ disk is occupied for the first time, and the magnetization doubles. This event is seen as a step of a factor of 2 in the specific heat.[12] We believe that the "layer promotion" events seen by Greywall at $n \gtrsim 0.240$ are these "disk occupation" events.

This model of the ^{3}He film admits a number of processes that lead to a magnetic interaction among the solid-layer particles. For example, an *in-plane* three-particle permutation, around the rudimentary triangle of the hexagonal lattice, leads to a ferromagnetic near-neighbor interaction[13] whose strength should be a very strong function of n_2. Greywall[4] finds n_2 to saturate at $n \simeq 0.25$ so that this interaction, while no doubt present, is not expected to be responsible for the strong dependence of the magnetic interaction on n at $n \gtrsim 0.25$ that he sees. Thus we seek an explanation of the exchange coupling at $n \gtrsim 0.25$ in terms of an RKKY mechanism involving the solid layer and the liquid layer.

An RKKY interaction results from a particle at site R of spin σ in the solid layer (R,σ) making a transition to the empty state (m',q',σ) in the liquid layer with a particle from an occupied state in the liquid layer (m,q,σ') going to (R,σ'). The intermediate state, a particle-hole pair, has energy $\epsilon_{m'q'} - \epsilon_{mq} > 0$. The further transition $(S,\sigma'') \rightarrow (m,q,\sigma'')$, $(m',q',\sigma) \rightarrow (S,\sigma)$ leads to exchange of the spins (σ,σ'') among the solid-layer sites (R,S) since $\sigma' = \sigma''$. This is a three-particle cyclic permutation. We have

$$H_x = \sum_{RS} K(R,S)\,\boldsymbol{\sigma}_R \cdot \boldsymbol{\sigma}_S\,, \tag{3}$$

with the strength of the exchange interaction given by

$$K(R,S) = \sum_{mm'qq'} \langle mq\sigma'', S\sigma \,|\, V \,|\, S\sigma'', m'q'\sigma\rangle \frac{1}{-\epsilon_{m'q'} + \epsilon_{mq}} \langle R\sigma', m'q'\sigma \,|\, V \,|\, mq\sigma', R\sigma\rangle N_{mq}(1 - N_{mq'})\,, \tag{4}$$

where N_{mq} is the Fermi-Dirac probability distribution and V the two-particle interaction. Because $K(R,S) < 0$ the interaction in Eq. (3) is ferromagnetic.

We focus on how $K(R,S)$ depends on the behavior of the liquid layer. Using Eqs. (1) and (2) and a contact interaction, $V \propto \delta(\mathbf{x} - \mathbf{x}')$, we can write $K(R,S)$ in the form

$$K(R,S) = \sum_{mm'qq'} \theta(\mu - \epsilon_{mq})\,\theta(\epsilon_{m'q'} - \mu) \frac{1}{-\epsilon_{m'q'} + \epsilon_{mq}} \times e^{i(\mathbf{q}-\mathbf{q}')\cdot(\mathbf{R}-\mathbf{S})} \,|\, A(\mathbf{q}-\mathbf{q}') \,|^2 \times |\, B_{mm'} \,|^2\,, \tag{5}$$

where

$$A(\mathbf{q}) = \int d\boldsymbol{\rho}\, e^{i\mathbf{q}\cdot\boldsymbol{\rho}} n_\parallel(\boldsymbol{\rho})\,,$$

$$B_{mm'} = \int dz\, \psi_m^*(z)\, n_\perp(z)\, \psi_{m'}(z)\,,$$

with

$$n_\parallel(\boldsymbol{\rho}) = |\, \psi_R(\mathbf{R} - \boldsymbol{\rho}) \,|^2, \quad n_\perp(z) = |\, \psi(z) \,|^2\,,$$

and μ is the Fermi energy of the particles in the liquid layer.[14] To get an idea of the content of Eq. (5) we look at the diagonal terms, $m = m'$, $K = \sum_m K_m$. For each disk the sum over q,q' gives a contribution that scales approximately as N_m, the number of particles in the disk.[15] Using the approximation

$$n_\perp(z) \propto \exp[-(z + z_0)/l]$$

we have

$$B_{mm} \propto \frac{1}{d} \frac{m^2}{m^2 + (d/\pi l)^2}\,. \tag{6}$$

For the factors in $K(R,S)$ that involve the liquid-layer

structure and the occupation numbers, we have

$$K(R,S) \propto \sum_m N_m \frac{1}{d}\left[\frac{m^2}{m^2+(d/2\pi l)^2}\right]^2. \quad (7)$$

This equation is an approximation to $K(R,S)$ as a sum of terms, one for each disk.[16] Each term involves exchange of a particle between two sites in the solid layer through "tunneling" out into the liquid, into the states $(m,\mathbf{q})$, and then back. The out and back process is controlled by B_{mm}. The motion of the particles parallel to the surface, going from **R** to **S** in the liquid, is controlled by N_m, a measure of the number of particles in the disk m.

In Fig. 3 we show $|K|^2$ vs d from Eq. (7). In constructing this figure we have used the ^{3}He film model of Guyer, McCall, and Sprague which provides a relationship between n_L, d, and the N_m. The values of $|K|^2$ in Fig. 3 have been normalized to 1 at large d, i.e., by the value of $|K|^2$ appropriate to the solid layer interacting with the bulk liquid. In this figure we also show the square of the coupling constant found by Greywall from an analysis of specific-heat data, his Fig. 27. We have normalized these data in the same way as above.[17] In Fig. 4 we show N_m vs d and K_m vs d for $m=1$–4 from the model of Guyer, McCall, and Sprague, and Eq. (7). Comparison of our result in Fig. 3 with Fig. 4 shows that oscillatory features in $|K|^2$ as d increases are due to the participation of additional disks in the exchange process. As d increases the contribution of each disk diminishes in a way that is compensated for by the increase in the number of participating disks.

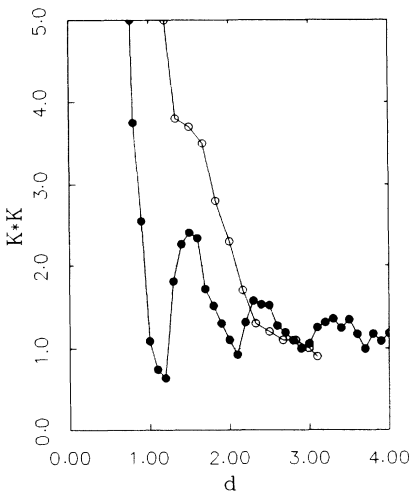

FIG. 3. Coupling constant. The value of the coupling constant from Eq. (7), squared, is plotted as a function of n_L (smooth curve). n_L is measured in layers; 1 layer = 0.7 atom/Å^2. Open circles are an experimental measure of the coupling constant squared from Fig. 27 of Greywall. The horizontal axis in this figure is the liquid-layer thickness in units of layers. For the data of Greywall use $d_3 = 2 + (n - 0.182)/0.70$ and $d = d_3 - 2$, so that fourth-layer promotion corresponds to $d \simeq 1$. Both curves have their magnitude scaled by the asymptotic value ($d \to +\infty$) to facilitate comparison and to remove factors from the theory that do not depend on n, e.g., the second-solid-layer wave function (Ref. 15).

In this paper we have argued for the extension to pure ^{3}He films of the physical picture developed by Guyer, McCall, and Sprague for mixture films. We take the qualitative and quantitative agreement between theory and experiment, Fig. 3, to provide strong support for this extension. To be fair, this conclusion is modestly circumscribed.

(1) There are "bumps" in the theory where none appear in the experiment. The reason for this difference could be that (a) the experimental substrate may be heterogeneous (see, for example, the explanation offered by Higley, Sprague, and Hallock for some features in their data) or (b) the step structure that is prominent in the magnetization is less so in the specific heat (the experimental points are from specific-heat data).[12]

(2) The magnitude of K^2 from Eq. (7) is in remarkable agreement with the magnitude of K^2 from experiment. They differ by a factor of about 2. This agreement should be regarded as fortuitous. "Exchange" interactions are notoriously hard to calculate. Agreement within an order of magnitude for K, let alone K^2, is often hailed. We have used a method of comparison of theory and experiment that removes some of the natural hazard often encountered. The degree to which we have been successful can only be tested by a much more substantial body of data. In the same vein we have made no attempt to correct our simple model for (a) interaction effects —as explicated, for example, by Krotschek, Saarella, and Epstein[18] or for (b) substrate scattering—as discussed by Tesanovic and co-workers.[19] Both can be im-

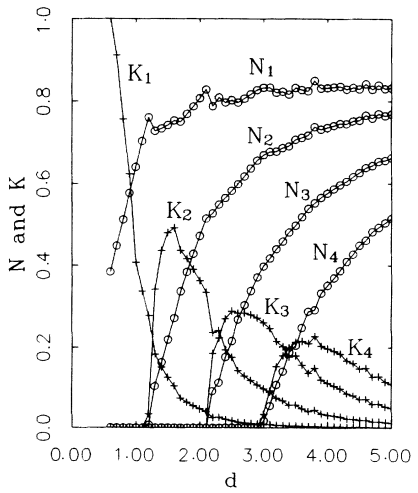

FIG. 4. N_m and K_m. The values of K_m from Eq. (7), for $m=1$–4, are plotted as a function d (smooth curves, arbitrary units). Shown also, by the circles, are $N_1, \ldots, N_4$ as a function of d.

portant and are the subject of our current research program. The experimental evidence, from Higley, Sprague, and Hallock is that there are important departures from our simple model that, while substantial, are not overwhelming.

The physical picture elaborated here provides a starting point from which to develop an understanding of the behavior of the two-dimensional neutral Fermi systems already studied by Greywall and by Higley, Sprague, and Hallock or yet to be studied on the (d_4,n) plane.

We thank D. T. Sprague and K. R. McCall for valuable discussions.

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[6]See, for example, M. Britz, J. G. Dash, D. C. Hickernell, E. O. McLean, and O. E. Vilches, Phys. Rev. A **8**, 1589 (1973); J. G. Dash, *Films on Solid Surfaces* (Academic, New York, 1974); S. W. Van Sciver and O. E. Vilches, Phys. Rev. B **18**, 285 (1978).

[7]As the areal density evolves, the film can be regarded as composed of several almost independent two-dimensional systems. Each system, a layer, has its own areal density. We quote areal densities in units of atom/Å^2 (usually the units are not displayed). At $n > 0.25$, the first layer is at high density, $n_1 \simeq 0.11^+$, and inert; the second layer is a low-density Fermi solid, $n_2 \gtrsim 0.05$; the third layer is a liquid layer of varying thickness, d. The liquid layers are described in terms of thickness in units of nominal layers, i.e., d/a_α, $a_\alpha = n_\alpha^{-1/3}$, $\alpha = 3,4$, where n_α is the bulk-liquid particle density.

[8]These ^{4}He layers hold the ^{3}He away from the strong van der Waals force of the substrate; the ^{3}He does not solidify. A small fraction of the ^{4}He is mobile and gives rise to a third-sound signal. See, for example, G. Agnolet, D. McQueeney, and J. D. Reppy, in *Proceedings of the Seventeenth International Conference on Low Temperature Physics, Karlsruhe, 1984,* edited by U. Eckern *et al.* (North-Holland, Amsterdam, 1984), Pt. 2, p. 965; J. Valles, thesis, University of Massachusetts (unpublished).

[9]R. A. Guyer, K. R. McCall, and D. T. Sprague, Phys. Rev. B **40**, 7417 (1989).

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[11]C. Kittel, in *Solid State Physics,* edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1969), Vol. 22, p. 1.

[12]The specific-heat "step" is more elaborate than its magnetization counterpart because of an additional contribution due to "promotion," i.e., dNm/dt.

[13]The triple exchange process is intrinsically ferromagnetic, see, e.g., L. Zane, Phys. Rev. Lett. **28**, 420 (1972). Since it involves the overlap of three pairs of localized wave functions, it is a very strong function of areal density.

[14]Except for the involvement of features that describe a thin film, $d,m,\ldots,$ the process we are describing is the same as one of several considered by H. Hecku and Y. Kuroda [Prog. Theor. Phys. **67**, 715 (1982)] for bulk ^{3}He.

[15]We have suppressed the weak dependence of the q,q' sum on several factors. For example, the involvement of $\exp[i(\mathbf{q}-\mathbf{q})\cdot(\mathbf{R}-\mathbf{s})]$. Numerical treatment of the double sum confirms weak dependence on this factor. We do not include a factor describing this dependence in Eq. (7). However, this dependence is included in the numerical determination of the coupling constant reported in Fig. 3. The role of the wave function describing the particles in the solid layer, i.e. $n_\parallel(\rho)$, is removed from the calculation by the "normalization" procedure discussed below Eq. (7). We do not need to make a specific choice for $n_\parallel(\rho)$.

[16]The off-diagonal terms, $m \neq m'$, are smaller than those we have included because of the energy denominator and interference effects. It is our intention to include such terms, along with a more elaborate treatment of the film structure problem, in future publications. We believe that the demonstration we are attempting in this paper is neither damaged nor saved by these terms.

[17]We use $d_3 = 2 + (n - 0.18)/0.07$ for $n \geq 0.18$ and $d = d_3 - 2$ in transferring the data of Greywall to Fig. 3.

[18]E. Krotschek, M. Saarella, and J. L. Epstein, Phys. Rev. Lett. **61**, 1728 (1988).

[19]See, for example, Z. Tesanovic, M. Jaric, and S. Mackawa, Phys. Rev. Lett. **57**, 2760 (1986).

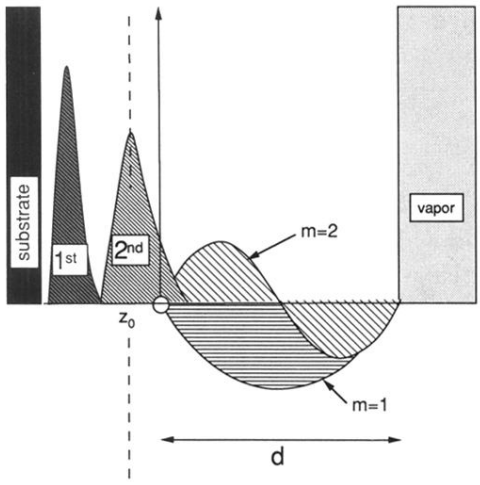

FIG. 2. Wave functions. At $n > 0.25$ atom/Å^2 the second solid layer ceases to evolve; the z part of the wave function of a particle in this layer, $\psi_\perp(z)$, sticks out into the liquid layer. In the liquid layer the particles are described by simple wave functions that evolve as n increases. The overlap integral between $\psi_\perp$ and the ψ_m leads to the n dependence of the RKKY coupling.