PHYSICAL REVIEW B VOLUME 40, NUMBER 4 1 AUGUST 1989

Equation of state for ^{3}He submonolayers physisorbed in ^{4}He-film surface states: Thermal Hartree-Fock approximation

R. H. Anderson
Department of Physics, Seattle Pacific University, Seattle, Washington 98119

M. D. Miller
Department of Physics, Washington State University, Pullman, Washington 99164-2814

(Received 1 February 1989)

A semiphenomenological effective interaction is introduced to describe ^{3}He physisorbed in ^{4}He-film surface states. The parameters in the effective interaction are fixed by requiring that the single-particle ^{3}He states are in agreement with available experimental data. The third-sound mediated effective ^{3}He-^{3}He interaction is then obtained in second-order perturbation theory (the one-ripplon exchange potential). The ^{3}He equation of state is computed using thermal Hartree-Fock for the one-ripplon exchange potential alone and with some account taken of the direct ^{3}He-^{3}He interaction. In no case do we find evidence of coexisting phases (van der Waals loops).

I. INTRODUCTION

The thin-film ^{3}He-^{4}He system has received extensive experimental and theoretical attention in the past few years. Notwithstanding, the present state of understanding is still muddled.

Theoretical treatments[1-3] of the two-dimensional ^{3}He-^{4}He mixture agree that at low enough temperatures, the system will phase separate. The phase separation in two dimensions will be complete rather than partial as in the bulk mixtures.[1] The reason for this difference is that, in two dimensions, ^{3}He is not a self-bound system. As discussed by Miller and Nosanow,[4] strictly two-dimensional ^{3}He is a gas at zero temperature. An indication of a possible phase separation in the mixture film was reported by Laheurte, Noiray, and Romagnan[5] who found anomalous low-temperature third-sound signals. The results, however, are apparently not reproducible.[6]

A layered phase separation has been seen in thicker films (5.7 layers where 1 layer = 3.6 Å) by Ellis, Hallock, Miller, and Guyer.[7] That is, the ^{3}He phase separates and floats on the superfluid ^{4}He film as a normal-fluid blanket. This layered configuration is driven by the van der Waals field of the film substrate. No onset has been observed for the transition and it is not clear whether the theoretical treatments of Refs. 1–3 can address this system in some film-averaged sense. The layered configuration has also been reported by Laheurte, Noiray, Romagnan, and Sornette[8] for this range of film thickness and temperature.

Bhattacharyya and Gasparini[9] (BG) reported apparent phase separation in mixture films of ≈ 4 layers of ^{4}He, submonolayer amounts of ^{3}He and $T < 100$ mK. BG analyzed heat capacities and argued that their low-temperature results are consistent with the formation of a condensed two-dimensional Fermi liquid. In very recent work, Valles, Higley, Johnson, and Hallock[10] reported that NMR measurements made on a mixture film system equivalent to that used by BG showed no apparent evidence for a phase transition.

In this paper, we shall attempt to construct a theoretical model to help guide the interpretation of the results of BG. The remarkable aspect of the BG results is the suggestion that the fermion-rich phase is self-bound. It is clear from Ref. 4, that if this is the case then the (^{4}He film) substrate must be playing a nontrivial dynamic role. In Sec. II, we shall develop a semimicroscopic description of the ^{3}He-^{3}He effective interaction mediated by the exchange of third-sound quanta. The approach is analogous to the phonon-mediated electron-electron interaction which is so familiar from solid-state physics. In Sec. III, we shall discuss the ^{3}He-substrate effective interaction which forms the basis of our numerical work. In Sec. IV, we compute the equation of state using thermal Hartree-Fock, and Sec. V is the Conclusion.

II. THE MODEL

We consider a film with N_4 ^{4}He atoms and N_3 ^{3}He atoms which is physisorbed to some solid substrate which occupies the lower ($z \leq 0$) half space. The ^{4}He film occupies area A and, in equilibrium, is laterally translationally invariant with film thickness d. (We use d to represent the height of the mobil layer above the solid surface as would be measured in a third-sound experiment.) The model Hamiltonian can be written

$$H = H_4 + H_3 + H_{34} \ , \tag{1}$$

where H_4 is the Hamiltonian of a pure ^{4}He film, H_3 is the Hamiltonian of the physisorbed ^{3}He, and H_{34} is the film-atom interaction. This starting point is familiar from many other systems, including electrons and phonons,[11] electrons on a ^{4}He substrate,[12] and spin-polarized hydrogen on a ^{4}He substrate.[13]

The ^{4}He-film Hamiltonian can be written

$$H_4 = \sum_k \hbar\omega_k (b_k^\dagger b_k + \tfrac{1}{2}) \ , \tag{2}$$

where k labels the momentum of a surface mode and the spectrum, ω_k, is given by[14]

$$\omega_k^2 = \left[\frac{v'_{s4}(d)}{m_4} + \frac{\sigma_0 k^2}{\rho_4} \right] q \tanh(qd) \ , \tag{3a}$$

$$q^2 = k^2 - \omega^2 / c_4^2 > 0 \ , \tag{3b}$$

where m_4 is the bare ^{4}He mass, $\rho_4 = m_4 n_4^0$ is the mass density, σ_0 is the zero-temperature surface tension and $v'_{s4}(z)$ is the substrate-film force. $v_{s4}(z)$ is taken to be of van der Waals form and so

$$v'_{s4}(d) = \frac{m_4 c_{3s}^2}{d} = \frac{3\alpha_s}{d^4} \ , \tag{4}$$

where α_s is the van der Waals parameter and c_{3s} is the third-sound speed. In Eq. (3b), c_4 is the bulk ^{4}He speed of sound. In the following, we shall treat the film as incompressible and so $q = k$. Below, we shall need the film fluctuation operator which can be written

$$\hat{\zeta}(\boldsymbol{\rho}) = \frac{1}{A^{1/2}} \sum_{\mathbf{k}} \left[\frac{\hbar k \tanh(kd)}{2\omega_k \rho_4} \right]^{1/2} e^{i\mathbf{k}\cdot\boldsymbol{\rho}} (b_{\mathbf{k}}^\dagger + b_{-\mathbf{k}}^\dagger) \ . \tag{5}$$

The Hamiltonian for a system of noninteracting physisorbed ^{3}He atoms can be written

$$H_3 = \sum_{\mathbf{k},\mu} \epsilon_{\mathbf{k},\mu} a_{\mathbf{k},\mu}^\dagger a_{\mathbf{k},\mu} \ , \tag{6}$$

where

$$\epsilon_{\mathbf{k},\mu} = \frac{\hbar^2 k^2}{2m_3^*} + \lambda_\mu \ , \tag{7}$$

and λ_μ is the eigenvalue of the Schrödinger equation:

$$-\frac{\hbar^2}{2m_3^*} \phi''_\mu(z) + v_3(z)\phi_\mu(z) = \lambda_\mu \phi_\mu(z) \ . \tag{8}$$

$v_3(z)$ is the single-particle effective interaction due to the substrate and film. A precise form for $v_3(z)$ will be discussed below. The eigenstates $\phi_\mu(z)$ represent the bound states in the ^{4}He film. In Eqs. (7) and (8), m_3^* is the ^{3}He effective mass which, in principle, is determined self-consistently from the Hamiltonian of Eq. (1). The calculation of m_3^* is discussed in detail in Sec. IV. In the numerical work which follows, we shall use $m_3^* = m_3$ the bare ^{3}He mass.

The Hamiltonian operator representing the interaction between the adsorbed quasiparticles and the modes of the film can be written

$$H_{34} = \int d\mathbf{r}_3 \int d\boldsymbol{\rho}_4 n_4^0 v_{34}(\boldsymbol{\rho}_3 - \boldsymbol{\rho}_4, z_3, z_4 = d) \times \hat{\psi}^\dagger(\mathbf{r}_3) \hat{\psi}(\mathbf{r}_3) \hat{\zeta}(\boldsymbol{\rho}_4) \ , \tag{9}$$

where n_4^0 is the bulk ^{4}He number density, v_{34} is an effective interaction between the ^{3}He and ^{4}He and $\psi(\mathbf{r})$ is a fermion destruction operator. The contribution to H_{34} representing the interaction of the ^{3}He and a static film has been neglected.

Now using

$$\hat{\psi}(\mathbf{r}) = \frac{1}{A^{1/2}} \sum_{\mathbf{k},\mu} e^{i\mathbf{k}\cdot\boldsymbol{\rho}} \phi_\mu(z) a_{\mathbf{k},\mu} \tag{10}$$

(where we have systematically suppressed the spin index since the interaction is spin independent), we obtain after some algebra

$$H_{34} = \frac{1}{A^{1/2}} \sum_{\substack{\mathbf{k},l \\ \mu,\nu}} (\Gamma_l^{\mu\nu} a_{\mathbf{k}+l,\mu}^\dagger a_{\mathbf{k},\nu} b_l + \Gamma_{-l}^{\mu\nu} a_{\mathbf{k},\mu}^\dagger a_{\mathbf{k}+l,\nu} b_l^\dagger) \ . \tag{11}$$

The vertex function $\Gamma_l^{\mu\nu}$ is given by

$$\Gamma_l^{\mu\nu} = \left[\frac{\hbar l \tanh(ld)}{2\omega_l \rho_4} \right]^{1/2} V_l^{\mu\nu} \ , \tag{12}$$

and $V_l^{\mu\nu}$ is the two-dimensional Fourier transform of the z-state matrix element of v_{34}:

$$V_l^{\mu\nu} = \int d\mathbf{s}\, e^{-l\cdot\mathbf{s}} \langle \mu | \mathbf{n}_4^0 v_{34}(s, z_3, z_4 = d) | \nu \rangle \tag{13}$$

and

$$\langle \mu | n_4^0 v_{34} | \nu \rangle = \int_{-\infty}^{\infty} dz \phi_\mu^*(z) n_4^0 v_{34}(s, z, z_4 = d) \phi_\nu(z) \ . \tag{14}$$

We can now obtain the third-sound mediated interaction from Eq. (11). The equation of motion for the fermion destruction operator can be written

$$-i\hbar \dot{a}_{\mathbf{k},\mu} + \epsilon_{\mathbf{k}} a_{\mathbf{k},\mu} = -\frac{1}{\sqrt{A}} \sum_{l,\nu} \Gamma_l^{\mu\nu} (b_l + b_{-l}^\dagger) a_{\mathbf{k}-l,\nu} \ . \tag{15}$$

Similarly, the equation of motion for the boson operator can be written

$$-i\hbar \dot{b}_{\mathbf{k}} + h\omega_k b_{\mathbf{k}} = -\frac{1}{\sqrt{A}} \sum_{\substack{\mathbf{m} \\ \mu,\nu}} \Gamma_{-\mathbf{k}}^{\mu\nu} a_{\mathbf{m}-\mathbf{k},\mu}^\dagger a_{\mathbf{m},\nu} \ . \tag{16}$$

We can now invert Eq. (16) and then pick off the interaction by inspection of Eq. (15). Introduce the propagator

$$id^0(\mathbf{k}, t; \mathbf{k}', t') = \langle 0 | T(b_{\mathbf{k}}(t) b_{\mathbf{k}'}^\dagger(t')) | 0 \rangle \ , \tag{17}$$

where T orders later times to the left and in the interaction representation

$$b_{\mathbf{k}}(t) = e^{-i\omega_{\mathbf{k}} t} b_{\mathbf{k}} \ . \tag{18}$$

The equation of motion for this propagator is

$$\left[i\frac{\partial}{\partial t} - \omega_k \right] d^0(\mathbf{k}, t - t') = \delta(t - t') \ . \tag{19}$$

Thus, Eq. (16) can be inverted by left multiplying by $d^0(\mathbf{k}, t - t')$ and integrating over t':

$$b_{\mathbf{k}}(t) = \frac{1}{A^{1/2}} \sum_{\substack{\mathbf{m} \\ \mu,\nu}} \left[\frac{\Gamma_{-\mathbf{k}}^{\mu\nu}}{\hbar} \right] \int dt' d^0(\mathbf{k}, t - t') \times a_{\mathbf{m}-\mathbf{k},\mu}^\dagger(t') a_{\mathbf{m},\nu}(t') \ . \tag{20}$$

We can proceed analogously for $b_{-\mathbf{k}}^\dagger$ and thus obtain

$$-i\hbar\dot{a}_{\mathbf{k},\alpha}+\epsilon_{\mathbf{k}}a_{\mathbf{k},\alpha} = -\frac{1}{A}\sum_{\substack{l,\mathbf{m}\\ \beta,\gamma,\delta}}\left[\frac{\Gamma_l^{\alpha\beta}\Gamma_{-l}^{\gamma\delta}}{\hbar}\right] \times\int dt'[d^0(l,t-t')+d^0(l,t-t')^*] \times[a^\dagger_{\mathbf{m}-l,\gamma}(t')a_{\mathbf{m},\delta}(t')a_{\mathbf{k}-l,\beta}(t)] \; . \tag{21}$$

Then, using $d^0(\mathbf{k},\omega)=(\omega-\omega_{\mathbf{k}}+i\eta)^{-1}$, the third-sound mediated interaction $\mathcal{V}_{\mathbf{k}}^{\alpha\beta\gamma\delta}(\omega)$ can be obtained by inspection of (21):

$$\mathcal{V}_{\mathbf{k}}^{\alpha\beta\gamma\delta}=\left[\frac{\Gamma_{\mathbf{k}}^{\alpha\beta}\Gamma_{-\mathbf{k}}^{\gamma\delta}}{\hbar}\right]\frac{2n_4^0 d\omega_k}{(\omega+i\eta)^2-\omega_k^2} \; . \tag{22}$$

III. THE EFFECTIVE INTERACTION

In his original treatment of ^{3}He surface states on a ^{4}He bulk planar surface, Lekner showed that the effective interaction seen by the ^{3}He atom has three basic features.[15] Within the bulk ^{4}He and away from the surface, the interaction goes to μ_3, the ^{3}He chemical potential. In the surface region there is a potential well preferentially occupied by the ^{3}He surface state. Finally, in the region outside of the ^{4}He and away from the surface the interaction goes to zero in a van der Waals manner.[16]

In a notable series of experiments, Gasparini and coworkers[17,18] have examined the behavior of the ^{3}He surface state in ^{4}He films by analyzing heat-capacity data. Their experiments revealed the presence of at least two bound states in films ≈ 4 layers thick. The system was reanalyzed by Sherrill and Edwards,[16] who showed that the ground-state eigenfunction was localized in the film surface and its eigenvalue was a monotonically increasing function of film thickness.

We shall model the ^{3}He-^{4}He effective interaction by a simple function of the form

$$v_{34}(\rho_{34},\, z_3,\, z_4)=\begin{cases}\infty, & z_3\le d_0\\[4pt] \dfrac{\mu_3}{n_4^0}\delta(\mathbf{r}_{34}), & d_0<z_3<d\\[8pt] -\dfrac{6\alpha_0}{\pi n_4^0(r_{34}^2+l^2)^3}, & d<z_3 \; .\end{cases} \tag{23}$$

The potential is infinite in the region of the high-pressure immobile first layer of ^{4}He. In the ^{4}He superfluid film, the interaction is a contact potential whose amplitude is fixed at the zero-concentration chemical potential for ^{3}He in bulk ^{4}He, μ_3, divided by the ^{4}He bulk density, n_4^0. In the region of the film surface and beyond, the potential is a soft-core van der Waals with the single adjustable parameter l determined by requiring that the bulk surface-state binding energy agrees with experiment. The quantity α_0 is the ^{4}He film-^{3}He van der Waals parameter. We shall now use v_{34} of Eq. (23) to determine $v_3(z)$ in Eq. (8) and $V_l^{\mu\alpha}$ in Eq. (13).

A. ^{3}He surface states

The ^{3}He atom feels a single-particle interaction $v_3(z_3)$ given by

$$v_3(z_3)=\int_{d_0}^{\infty}dz_4\int d\rho_{34}n_4(z_4)v_{34}(\rho_{34},z_3,z_4) +\int_{-\infty}^{0}dz_s\int d\rho_{s3}n_s v_{s3}(r_{s3})[1-f(z_3-d)] \; , \tag{24}$$

where v_{s3} is the substrate-helium van der Waals interaction, and $f(z-d)$ is the film surface profile normalized to unity,

$$f(z-d)=n_4(z)/n_4^0 \; . \tag{25}$$

This function is used to distinguish between inner-film quantities and surface quantities. For example, the substrate potential in Eq. (24) only explicitly contributes in the outer-film region. This is to prevent the substrate potential from attracting all the ^{3}He atoms into the immediate vicinity of the substrate due to the absence of short-range correlations in this theory. In the physical system the substrate potential produces a small density gradient in the ^{4}He film which prevents the ^{3}He from accumulating near the substrate.

For the film density profile, we use the form introduced by Mantz and Edwards as discussed in Ref. 19. Thus,

$$f(z-d)=1/(1+e^{p(z)})^2 \; , \tag{26}$$

where

$$p(z)=\beta_1(z-d)+\beta_2+\beta_3/[(z-d)^2+\beta_5]+\beta_4/(z^2+\beta_5) \; . \tag{27}$$

The values for the β_i and all the parameters used in $v_3(z)$ are given in Table I.

In the approximation where $n_4(z)$ in Eq. (24) is replaced by a step function, $n_4(z)=n_4^0\theta(d-z)$, the integrals can be carried out analytically to yield

$$v_3(z_3)=\left[-\frac{\alpha_s}{z_3^3}-\frac{3}{2}\left[\frac{\alpha_0}{l^3}\right]\{(\theta_d-\theta_0) -\tfrac{1}{2}[\sin(2\theta_d)-\sin(2\theta_0)]\}\right] \times[1-f(z_3-d)]+\mu_3 f(z_3-d) \; , \tag{28}$$

TABLE I. Values of the parameters used in the effective interaction.

Parameter	Value
μ_3	2.785 K
n_4^0	0.0218 Å^{-3}
σ_0	0.278 K/Å^2
α_0	120 K Å^3
α_s	1,740 K Å^3
β_1^2	$(1.1816+267/d^3)$ Å^{-2}
β_2	-2.5
β_3	$(20/4\beta_1)$ Å^{-2}
β_4	$(267/4\beta_1)$ Å^{-2}
β_5	72.25 Å^{-2}
l	2.88 Å

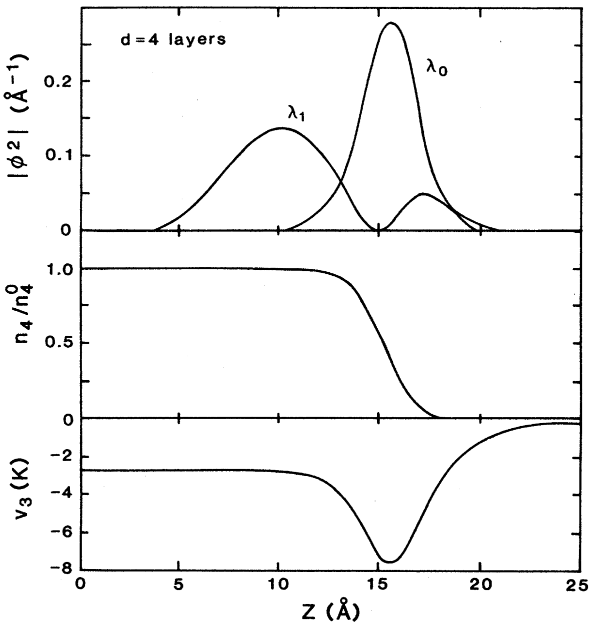

FIG. 1. The probability densities of the ground and first excited surface state for ^{3}He in a $d=4$ layer ^{4}He film. Also shown are the ^{4}He surface profile from Eq. (26) and the ^{3}He single-particle interaction, Eq. (24).

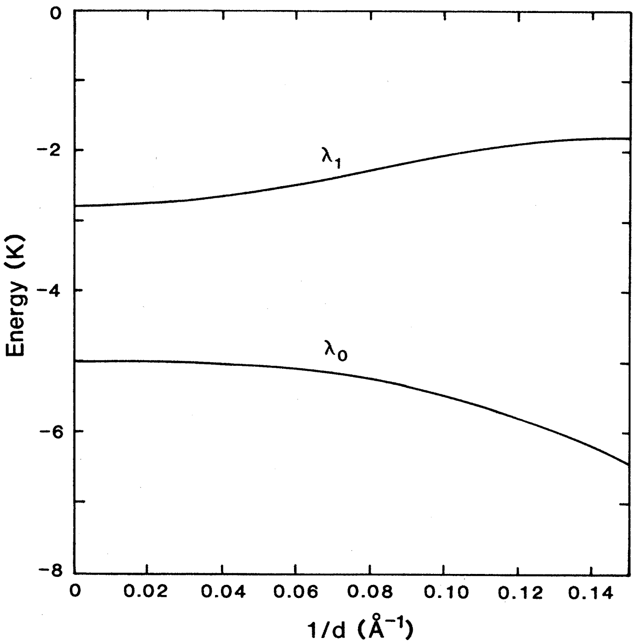

FIG. 2. The ground state and first excited state eigenvalues as a function of inverse ^{4}He film thickness.

where α_s is the substrate-^{3}He van der Waals parameter as in Eq. (4), and

$$\theta_0 \equiv \tan^{-1}\left[\frac{l}{z_3-d_0}\right],$$

$$\theta_d \equiv \tan^{-1}\left[\frac{l}{z_3-d}\right].$$

For the step-function model, Eq. (28), $l=2.64$ Å as compared with $l=2.88$ Å for the model with a realistic surface profile.

The potential, $v_3(z)$, together with the ground state and first excited state probability densities are shown in Fig. 1 for a film with $d=14.4$ Å (4 layers). These results are in good agreement with the calculation of Sherrill and Edwards.[19] In Fig. 2, we show the two lowest eigenvalues as a function of film thickness. These results indicate that the ground-state eigenvalue, λ_0, is a monotonically increasing function of film thickness (the decrease in λ_0 with decreasing film thickness is due simply to the increasing importance of the substrate van der Waals field on the surface state). The first excited state, λ_1, can undergo an interesting maximum as a function of film thickness as can be seen in Fig. 2 of Ref. 19. The maximum is due to a crossover from the large d region, where λ_1 is dominated by the gain in kinetic energy with decreasing film thickness, to a small d region, where λ_1 is dominated by the decrease in potential energy with decreasing film thickness. As seen in Fig. 2, the maximum occurs in this model for a very thin film. [The maximum for the step-function model, Eq. (34), occurs at $d\approx 9$ layers since in this model the substrate potential plays a slightly greater role in the surface region.] In summary, we have determined $l=2.88$ Å by requiring that $v_3(z)$ has the correct *bulk* surface-state binding energy and we then demonstrated that our simple model gives good agreement with the calculations of Refs. 17 and 19 for the film states.

B. Effective interaction

We first compute the vertex function, Eq. (12). Substituting Eq. (23) into Eq. (13), we find

$$V_{\mathbf{k}}^{\mu\nu}=\mu_3\phi_\mu^*(d)\phi_\nu(d)-\tfrac{3}{4}\alpha_0 k^4\left\langle\mu\left|\left[\frac{K_{-2}(k[(z-d)^2+l^2]^{1/2})}{(k[(z-d)^2+l^2]^{1/2})^2}\right][1-f(z-d)]\right|\nu\right\rangle . \tag{29}$$

where K_{-2} is a modified Bessel function. The $k\rightarrow 0$ limit of Eq. (29) is of interest. Using the asymptotic form[20]

$$K_{-2}(x)\approx 2/x^2-(x^2/8)\ln(x/2)+\cdots ,$$

we find

$$V_0^{\mu\nu}=\mu_3\phi_\mu^*(d)\phi_\nu(d)$$
$$-\tfrac{3}{2}\alpha_0\langle\mu|[(z-d)^2+l^2]^{-2}[1-f(z-d)]|\nu\rangle , \tag{30}$$

and $V_0^{\mu\nu}$ is clearly nonzero. If we write the vertex functions as

$$\Gamma_k^{\mu\nu} = C_k V_k^{\mu\nu} , \tag{31}$$

where from Eq. (12)

$$C_k = \left[\frac{\hbar k \tanh(kd)}{2\omega_k \rho_4} \right]^{1/2} , \tag{32}$$

then, from Eq. (3a), $\lim_{k \to 0} \omega_k \equiv c_{3s} k$; thus

$$\lim_{k \to 0} C_k \equiv \left[\frac{\hbar d}{2\rho_4 c_{3s}} k \right]^{1/2} . \tag{33}$$

Therefore,

$$\lim_{k \to 0} \Gamma_k^{\mu\nu} \sim O(k^{1/2} d^{5/4}) . \tag{34}$$

The vertex function vanishes like the square root of the wave vector and increases with increasing film thickness (the implicit d dependence in the states ϕ_μ^* and ϕ_ν has been neglected).

In the equation-of-state calculations to be discussed in the next section, we shall confine ourselves to temperatures and densities low enough so that the excited z states, λ_1, etc., are negligibly occupied. We shall therefore only consider the ground-state matrix element $\mu, \nu = 0$ and for clarity suppress the $\mu\nu$ superscripts in Γ and the effective interaction henceforth.

In Fig. 3, we examine V_k for $d = 4$, 6, 8, and 10 layers. The function becomes less negative with increasing film thickness; however, the dependence on d is very mild. The large k asymptote is given by the contact interaction contribution and is ~ -0.6 K. In Fig. 4 we show the vertex functions Γ_k for the same systems as in Fig. 3, $d = 4$, 6, 8, and 10 layers. The coupling function C_k, Eq. (32), forces the Γ_k to zero as $k \to 0$ with a square-root-type cusp as shown in Eq. (34). In the limit $kd \gg 1$, $C_k \sim O(k^{-1/4})$ and Γ_k vanishes very slowly. The excitation spectra ω_k are shown in Fig. 5 [cf. Eqs. (3a) and

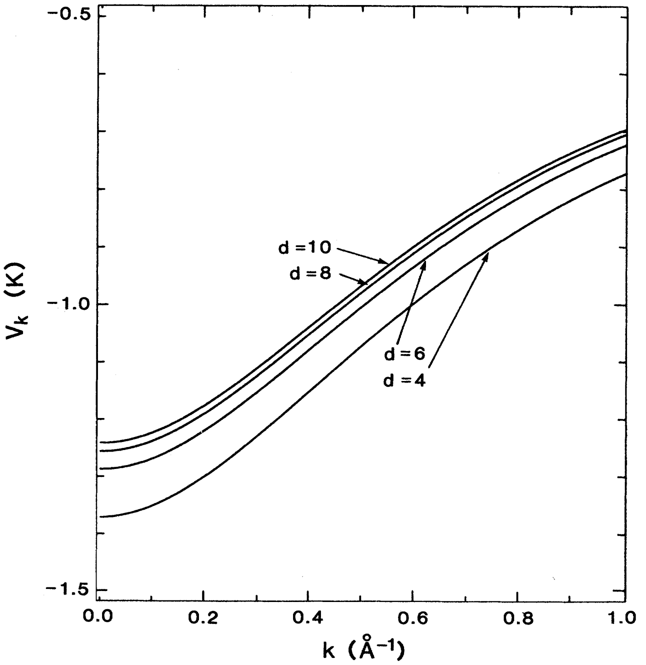

FIG. 3. The function V_k from Eq. (29) for ^{4}He films with thickness $d = 4$, 6, 8, and 10 layers.

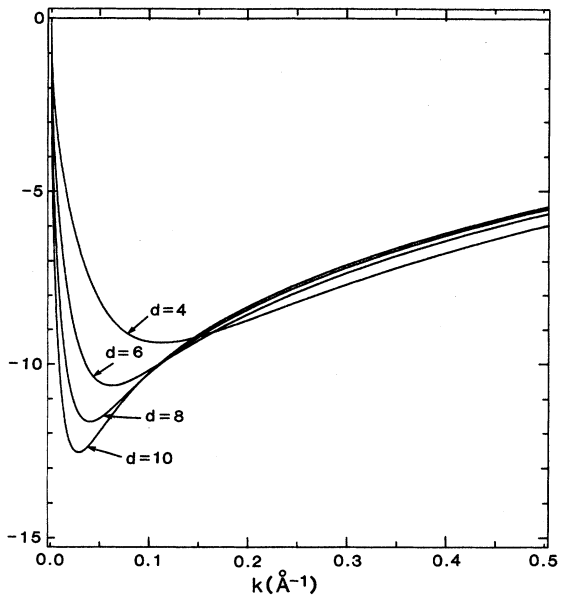

FIG. 4. The vertex function Γ_k from Eq. (12) for ^{4}He films with thickness $d = 4$, 6, 8, and 10 layers. As shown in Eq. (34) Γ_k vanishes like $k^{1/2}$ as $k \to 0$.

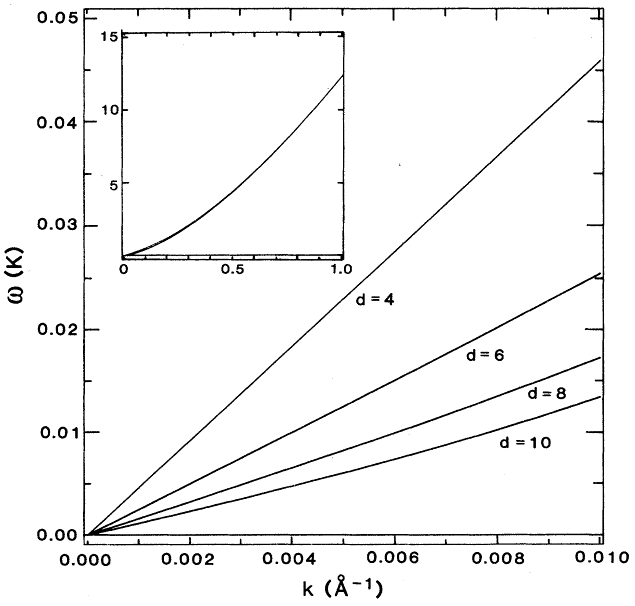

FIG. 5. The ^{4}He-film excitation spectra from Eqs. (3a) and (3b). The softening in the spectrum with increasing d is only evident at very small values of k as shown by the inset.

(3b)]. There is a crossover wave vector, $k_c \equiv (3\alpha_s n_4^0/\sigma_0)d^{-2}$, such that for $k < k_c$ ($> k_c$) the spectrum is dominated by the substrate (surface tension) restoring force. For $d=4$ layers, $k_c \approx 0.01$ Å^{-1}, thus the spectra are basically identical (i.e., independent of d) for $k > 0.01$ Å^{-1} for the systems we have considered in Figs. 3 and 4 above. The spectra out to 2 Å^{-1} are shown in the inset to Fig. 5, whereas Fig. 5 concentrates on the $k < k_c$ region. The softening of the small k spectrum with increasing film thickness is evident.

In the static limit, the *one-ripplon exchange potential* (OREP) from Eq. (22) can be written

$$\mathcal{V}_{\mathbf{k}} = -\frac{2n_4^0 d|\Gamma_k|^2}{\hbar\omega_k} . \tag{35}$$

In Fig. 6, we show $\mathcal{V}_{\mathbf{k}}$ for the systems $d=4$, 6, 8, and 10 layers. The OREP is manifestly negative as is to be expected from second-order perturbation theory. In the limit $k \to 0$, it is straightforward to show that

$$\lim_{k\to 0} \mathcal{V}_{\mathbf{k}} = -\frac{(V_0 d)^2}{m_4 c_{3s}^2} \sim O(d^5) , \tag{36}$$

if we ignore d dependence in the matrix element V_0. Thus, the interaction becomes long ranged in real space as d gets large. $\mathcal{V}_k$ is very short ranged in k space (e.g., the $d=10$ potential increases from $-18\,000$ K at $k=0$ to -1.4 K at $k=1$ Å^{-1}).

In the following section, we shall utilize the OREP in a thermal Hartree-Fock approach in order to compute the ^{3}He equation of state. In addition, we shall augment the OREP with two models to account for the direct ^{3}He-^{3}He interaction and the induced ^{3}He-^{3}He interaction due to the ^{4}He background.

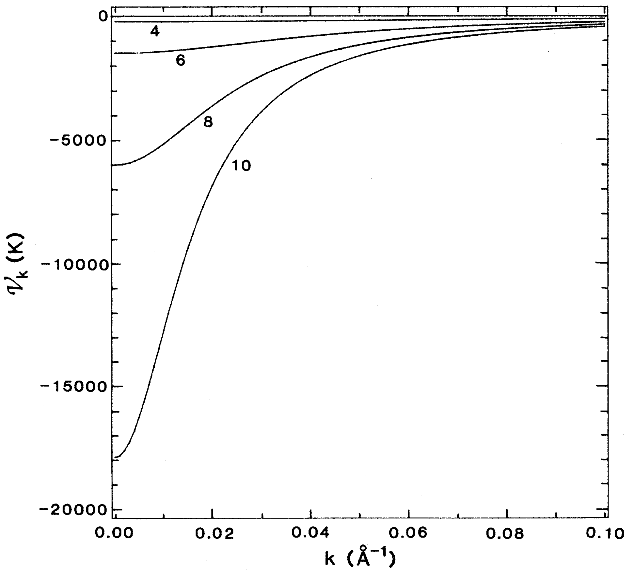

FIG. 6. The one-ripplon exchange potential, $\mathcal{V}_{\mathbf{k}}$, from Eq. (35) for ^{4}He films of thickness $d=4$, 6, 8, and 10 layers. The long-ranged behavior as d increases is evident [cf. Eq. (36)].

IV. THERMAL HARTREE-FOCK

As discussed by Mermin[21] and des Cloizeaux,[22] one can readily generalize the usual zero temperature, variational, mean-field Hartree-Fock theory to finite temperatures. Thermal Hartree-Fock (THF) is the natural place to begin in describing a system like adsorbed ^{3}He interacting weakly through an OREP since the usual problem with HF, its inability to deal with strong short-ranged correlations, is of no consequence.

From Ref. 22, the (spreading) pressure can be written

$$P = (2s+1)\int \frac{d\mathbf{k}}{(2\pi)^2} n_{\mathbf{k}} \Sigma_{\mathbf{k}}^* + \frac{(2s+1)}{\beta}\int \frac{d\mathbf{k}}{(2\pi)^2} \ln(1+e^{-\beta(\epsilon_{\mathbf{k}}-\mu)}) , \tag{37}$$

where $s=\frac{1}{2}$ for ^{3}He, $\Sigma_{\mathbf{k}}^*$ is the proper self-energy in the Hartree-Fock approximation,[23]

$$\Sigma_{\mathbf{k}_1}^* = \int \frac{d\mathbf{k}_2}{(2\pi)^2} [(2s+1)V_0 - V_{\mathbf{k}_{12}}] n_{\mathbf{k}_2} , \tag{38}$$

$n_{\mathbf{k}}$ is the average occupation number,

$$n_{\mathbf{k}} = \frac{1}{e^{\beta(\epsilon_{\mathbf{k}}-\mu)}+1} , \tag{39}$$

and

$$\epsilon_{\mathbf{k}} = \epsilon_{\mathbf{k}}^0 + \Sigma_{\mathbf{k}}^* , \tag{40}$$

where $\epsilon_{\mathbf{k}}^0$ is the free-particle spectrum, β is the inverse temperature and μ is the chemical potential. Thus, for a given density and temperature one can self-consistently solve Eqs. (38)–(40) for the self-energy and chemical potential. These quantities then yield the equation of state

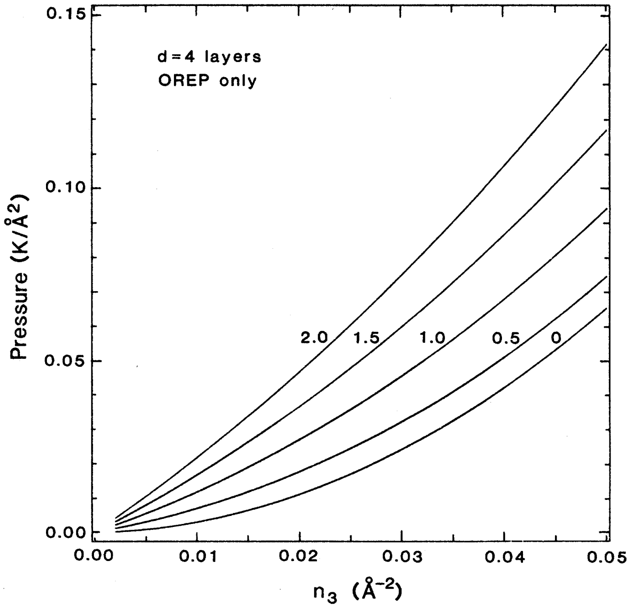

FIG. 7. The equation of state of adsorbed ^{3}He in the thermal Hartree-Fock approximation using only the OREP, Eq. (35). Isotherms at $T=0$, 0.5, 1.0, 1.5, and 2.0 K are shown. There is no evidence for coexisting phases.

from Eq. (37).

It is important to point out that, for this system, there are no so-called tadpole diagrams[24] which correspond to the direct term in Eq. (38) [i.e., $V(k\equiv 0)=0$]. Physically, we certainly do not expect that a uniform displacement of the surface could lead to an enhanced interaction. A rigorous discussion of this point can be found in the Appendix.

In the following we shall present three calculations of ^{3}He equations of state. The first equation of state shall be computed using only the OREP. In the second, we shall incorporate some features of the direct ^{3}He-^{3}He interaction by using a hard-core form for the ^{3}He chemical potential. Third, we shall adapt the bulk ^{3}He-^{3}He effective interaction as introduced by Owen[25] to the surface mixture. We shall search for condensation by looking for the appearance of van der Waals loops as is typical of mean-field theories.

A. OREP potential

In Fig. 7, we show the pressure as a function of density at a series of temperatures between 0 and 2 K. The equations of state are concave functions. There is no evidence of coexisting phases.

The OREP used for these equations of state is temperature and concentration dependent through the third-sound spectrum. Any dependence which can cause the spectrum to soften can have an important influence on the magnitude and range of the potential, Eq. (35). For example, Eq. (4) has a coefficient of $n_s(T)/n_4$, the superfluid fraction. As $T\rightarrow T_\lambda$, $n_s(T)\rightarrow 0$, and so the spectrum softens as the temperature is raised. We model this temperature dependence by extrapolating the low-temperature phonon dominated behavior to higher temperatures:[26]

$$\frac{n_s(T)}{n_4}=1-\left[\frac{T}{T_\lambda}\right]^4 . \tag{41}$$

The superfluid density is only mildly temperature dependent until temperatures approach the region of the λ point. However, since our theory is in any case inapplicable in that region, we feel Eq. (41) is sufficiently accurate for our purposes.

We have also included the $T^{7/3}$ ripplon contribution to the ^{4}He surface tension.[27] Thus, the surface tension in Eq. (3a) is replaced by

$$\sigma(T)=\sigma_0-(4.72\times 10^{-3})T^{7/3} , \tag{41a}$$

where T is in K and the surface tension is in K/Å^2. There are also less important third-sound and ^{3}He-concentration effects which have been omitted.

B. Hard core

An approximation of the effects of a hard core in the ^{3}He-^{3}He interaction can be incorporated into the THF approximation by demanding that the chemical potential be determined by

$$\frac{n_3}{1-n_3a_c}=\frac{1}{N_3}\sum_{\mathbf{k}} n_{\mathbf{k}} , \tag{42}$$

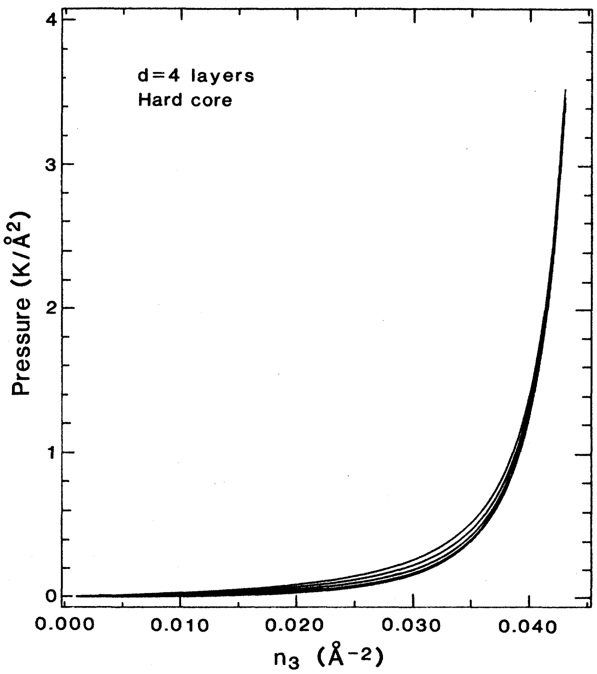

FIG. 8. The equation of state of adsorbed ^{3}He in the thermal Hartree-Fock approximation using the OREP, Eq. (35), and a chemical potential determined with an excluded area, Eq. (42). There is no evidence for coexisting phases.

where a_c is a hard-core excluded area. For definiteness, we set $a_c=\pi\sigma_{\rm LJ}^2$ where $\sigma_{\rm LJ}=2.556$ Å is the Lennard-Jones length parameter. The equation of state, Eq. (36), is computed after solving Eqs. (37) and (42) self-consistently. The results are shown as Fig. 8.

The effect of the hard-core repulsion on the pressure is evident. However, there is, once again, no evidence for van der Waals loops and coexisting phases. The system in Fig. 8 has $d=4$ layers and on this scale of pressures, differences in the isotherms from $T=0$ K to $T=2$ K are barely discernible.

C. Owen's effective interaction

Using a variational ansatz, Owen[25] was able to write down an effective interaction for two ^{3}He atoms in a bulk ^{4}He system. In the following we shall make a simple adaptation of his results to the surface system.

Owen showed that the ^{3}He-^{3}He interaction due to the direct (Lennard-Jones) interaction plus the induced interaction due to the ^{4}He background could be written

$$W_{33}(r)=g_{33}(r)[v_{33}(r)+t(r)+w(r)] , \tag{43}$$

where $v_{33}(r)$ is the direct (Lennard-Jones) interaction,

$$t(r)=\frac{\hbar^2}{m_3}\left[\frac{d}{dr}(g_{33}(r))^{1/2}\right]^2 , \tag{44}$$

is the correlational kinetic energy and the Fourier transform of $w(r)$ is

$$\tilde{w}(k) = -\left[\frac{\hbar^2 k^2}{4m_4 n_4^0 S_{44}^2(k)}\right][S_{34}(k)-1]^2 \times \left[\frac{2m_4}{m_{34}} S_{44}(k) - 1\right] . \qquad (45)$$

$S_{\alpha\beta}(k)\alpha,\beta=3,4$, the liquid structure factors, are the Fourier transforms of the radial distribution functions $g_{\alpha\beta}(r)$. Here,

$$\frac{1}{m_{34}} = \tfrac{1}{2}(1/m_3 + 1/m_4)$$

and for definitions of the distribution functions see, for example, Ref. 1.

The numerical work was carried out using a parametrized product of pair functions as described in Ref. 28. We substituted the Jackson-Feenberg form for the correlational kinetic energy, Eq. (44), and augmented the small-k behavior of $S_{44}(k)$ and $S_{34}(k)$ to agree with experiment. We find excellent agreement between our $\tilde{w}(k)$ and the results of Owen.

We can now model the surface system as a uniform slice of bulk by identifying the Fermi circle of a two-dimensional surface system as a great circle of the Fermi sphere of a model three-dimensional system. Thus we find

$$n_{3b} = \tfrac{4}{3}(2n_3^3/\pi)^{1/2} , \qquad (46)$$

where n_{3b} is the number density of the model bulk system. We note that Eq. (45) was derived in the zero-^{3}He concentration limit; however, the effective interaction as implemented in this work depends indirectly on n_3 through the distribution functions $g_{\alpha\beta}$.

Thus, within our model, the surface equivalent of Owen's effective interaction can be obtained from

$$\mathcal{W}_{\mathbf{k}} = \int d\mathbf{s} e^{i\mathbf{k}\cdot\mathbf{s}} W_{\mathrm{eff}}(s) , \qquad (47)$$

where **k** and **s** are two-component vectors and

$$W_{\mathrm{eff}}(s) = \int_{-\infty}^{\infty} dz \int_{-\infty}^{\infty} dz' n_3(z) n_3(z') W_{33}(s, z-z') . \qquad (48)$$

In Fig. 9, we illustrate $\mathcal{W}_{\mathbf{k}}$ for $d=4$ layers and $n_3=0.07$ Å^{-2}. This interaction can be added directly to $\mathcal{V}_{\mathbf{k}}$, the OREP, in Eq. (37) for the self-energy. Then once more solving Eqs. (38)–(40) self-consistently, we obtain the equation of state in the THF approximation. In Fig. 10, we show the results of these computations for $d=4$ layers. The slight rippling at high density is artificial and can be removed by utilizing a finer grid in the parameters for the radial distribution functions.

The isotherms are once more monotonic with no evidence of van der Waals loops. The isotherms appear similar to ideal fermi-gas isotherms. In order to display clearly the influence of interactions, we show in Fig. 11 the chemical potential difference between the computed system and an ideal Fermi gas at the same temperature and density. The temperature dependence is clearly much weaker than the density dependence.

The ^{3}He surface effective mass can be obtained from Eq. (40):

$$\frac{1}{m^*} = \frac{1}{\hbar^2 k_F}\left[\frac{d\epsilon_k}{dk}\right]_{k_F}$$

or (49)

$$\frac{m}{m^*} = 1 + \frac{m}{\hbar^2 k_F}\left[\frac{d\Sigma_{\mathbf{k}}^*}{dk}\right]_{k_F} .$$

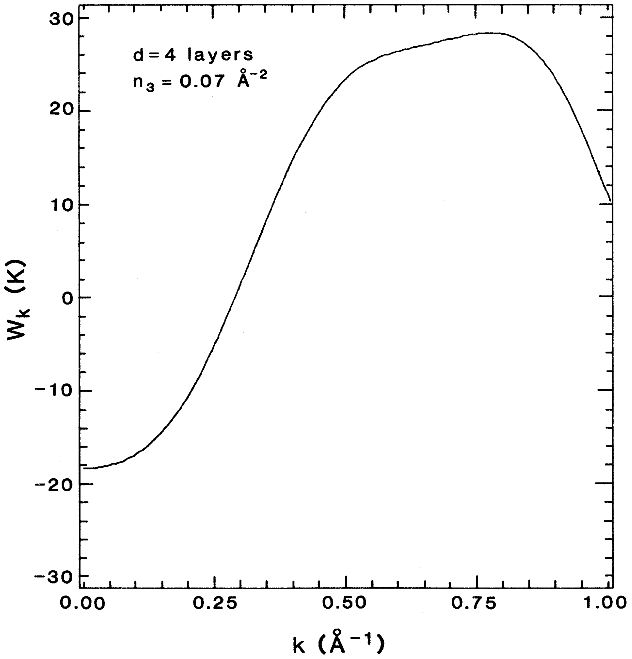

FIG. 9. Owen's effective interaction from Eqs. (43)–(48). The results shown are for a ^{4}He film with thickness $d=4$ layers and adsorbed ^{3}He at density $n_3=0.07$ Å^{-2}.

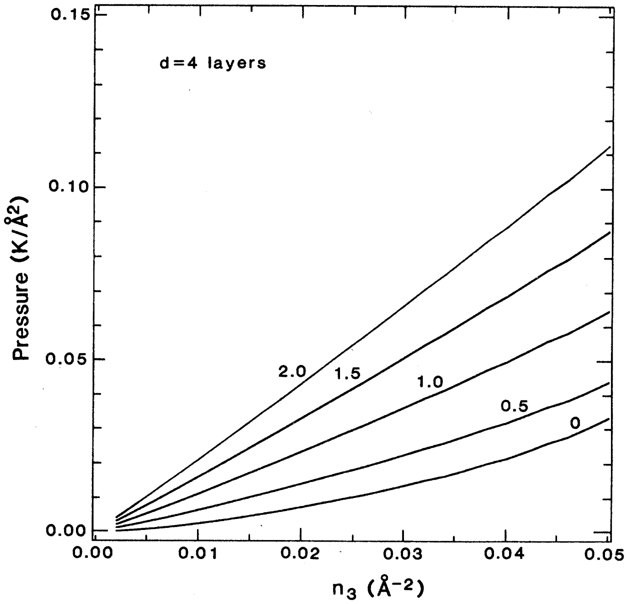

FIG. 10. The equation of state of adsorbed ^{3}He in the thermal Hartree-Fock approximation using the OREP, Eq. (35), and Owen's effective interaction, Eqs. (43)–(48). There is no evidence for coexisting phases.

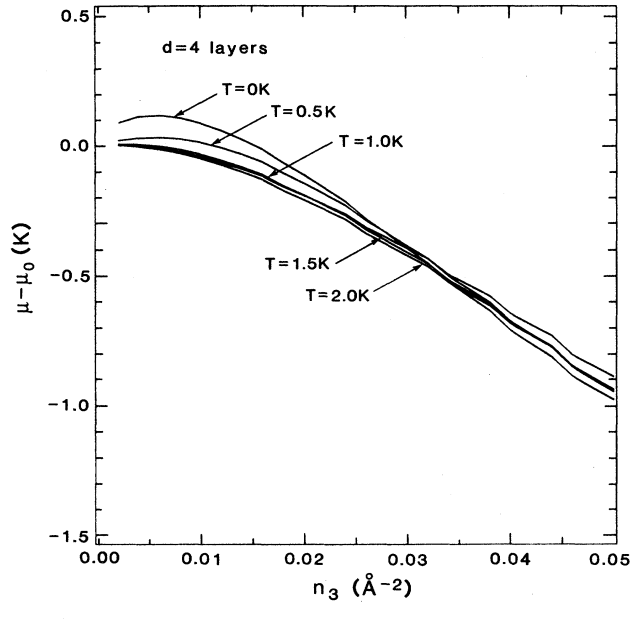

FIG. 11. The chemical potential differences between the adsorbed ^{3}He and an ideal two-dimensional Fermi gas showing the influence of the attractive effective interactions. The data shown are at $T=0$, 0.5, 1.0, 1.5, and 2.0 K. The $T=0$ K result is the maximum positive curve at small n_3.

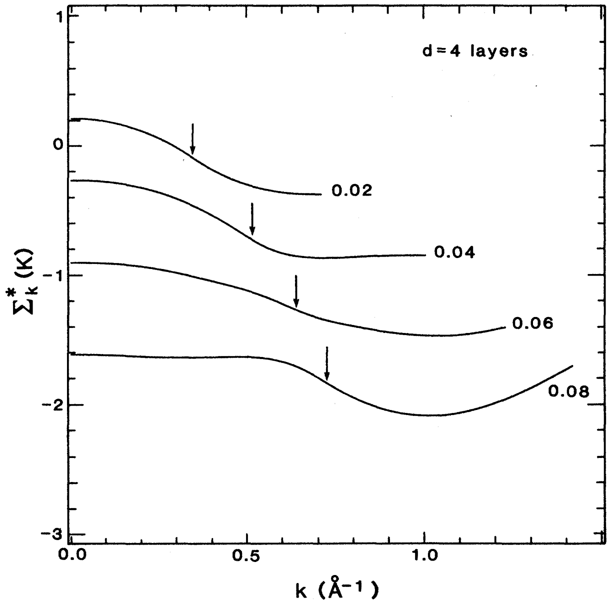

FIG. 13. The proper self-energy for ^{3}He systems with $n_3=0.02$, 0.04, 0.06, and 0.08 Å^{-2} on a $d=4$ layer ^{4}He film. The vertical arrows mark the positions of the Fermi momenta.

In Fig. 12, we show m/m^* as a function of n_3 for $d=4{,}8$ layers. In recent work, Wang and Gasparini[29] have reported measurements of the ^{3}He effective mass in ^{4}He surface states. For $d=4$ layers and $n_3=0.02$ Å^{-2}, we find $m/m^*=1.45$ which is in reasonable agreement with ~1.55 reported in Ref. 29. The results in Fig. 12 also indicate that m/m^* monotonically increases with increasing n_3 (Wang and Gasparini do not report density dependencies for the effective mass). The effective mass of the $d=8$ layer system is negative at small densities, a result which we attribute to the breakdown of the applicability of the OREP as will be discussed in the next section.

It is useful to examine Σ_k^* for the $d=4$ and 8 layer systems in order to see the basis for the differences in the effective masses. In Figs. 13 and 14, we show Σ_k^* for $d=4$ and 8 layers, respectively. The four results are for

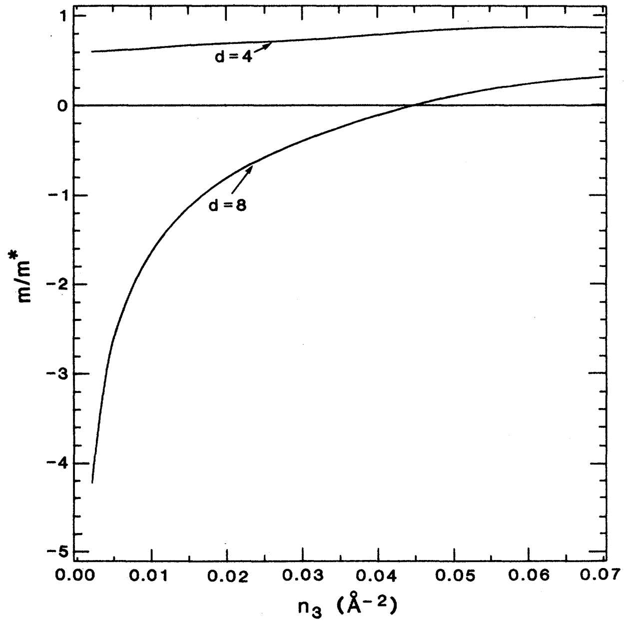

FIG. 12. The ^{3}He effective mass as a function of areal density for ^{4}He films with $d=4$ and 8 layers.

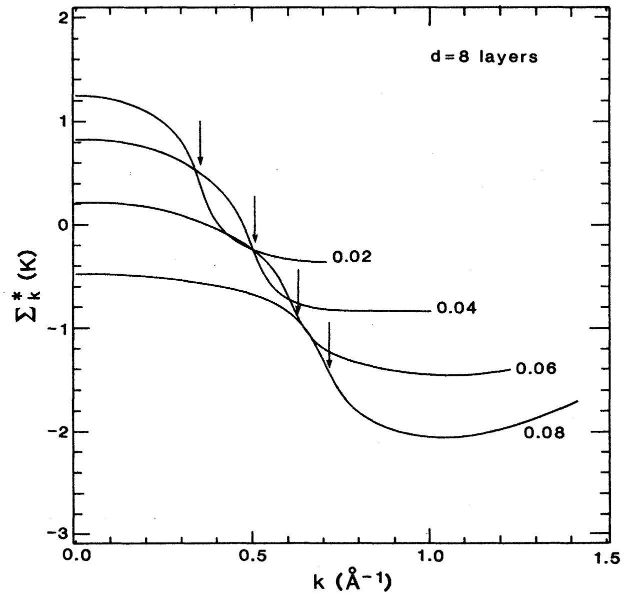

FIG. 14. The proper self-energy for ^{3}He systems with $n_3=0.02$, 0.04, 0.06, and 0.08 Å^{-2} on a $d=8$ layer ^{4}He film. The vertical arrows mark the positions of the Fermi momenta.

$n_3=0.02$, 0.04, 0.06, and 0.08 Å^{-2}. The positions of the Fermi wave vector ($k_F^2=2\pi n_3$) are denoted by arrows. The slopes at $k=k_F$ are all negative and so $m/m^* < 1$.

V. CONCLUSION

In this paper we have computed in second-order perturbation theory the ^{3}He-^{3}He effective interaction due to the exchange of third-sound quanta. The equations of state computed in the THF approximation show no van der Waals loops.

The limits of validity of the theory can be argued as follows. The ^{3}He surface states are computed with a model in which the ^{4}He superfluid film is treated as a uniform slice of the bulk. Thus the theory is not applicable to ^{3}He in low-thickness films where Krotscheck[30] showed that there was an appreciable structure normal to the substrate. The theory is also not applicable in the limit of large-thickness films as the induced interaction $\mathcal{V}_k$, Eq. (35), becomes too attractive. Krotscheck, Saarela, and Epstein[31] have argued that the proper d dependence in the matrix element Γ_k cures that problem.

In very recently published work, Krotscheck, Saarela, and Epstein[32] have reported calculations of the ^{3}He-^{3}He effective interaction in ^{4}He films. Their calculations proceeded from a variational, microscopic point of view with no adjustable parameters. Our adaptation of the Owen interaction, Fig. 9, is in good, qualitative agreement with their results. The basic difference is their result has its first maximum at $k \sim 1$ Å^{-1}, whereas ours has its first maximum at $k \sim 0.8$ Å^{-1}.

Our approach is semiphenomenological in that we take advantage of some of the known experimental results in order to determine our wave function as compared to the correct but much more complicated task of Krotscheck *et al.*[31,32] of computing everything from a given Hamiltonian. In further work, we shall amend our theory to give proper d dependence and shall then go beyond Hartree-Fock to compute the equations of state.

ACKNOWLEDGMENTS

One of the authors (R.H.A.) wishes to express appreciation to E. K. Riedel for originally suggesting the problem and to J. G. Dash, M. J. Schick, and C. J. Pethick for interesting and helpful discussions. Both authors acknowledge the support of the Research Corporation.

APPENDIX

In the text the tadpole diagram was ignored on the basis of physical argument. The vanishing of the tadpole diagram can also be shown analytically from the momentum dependence of the ^{3}He-ripplon vertex as calculated from Eq. (9). When the integration in that equation is carried out with respect to $\boldsymbol{\rho}_3-\boldsymbol{\rho}_4$ and $\boldsymbol{\rho}_4$ the conservation of film mass requires that

$$\int d^2\rho_4 \zeta(\rho_4)=0 ,$$

since $\zeta(\rho_4)$ is the deviation of the surface from the equilibrium configuration at position $\boldsymbol{\rho}_4$. Consequently for lateral momentum

$$\Gamma_0^{\mu\nu}=0 .$$

Equation (16) implies, therefore, that b_0 is a constant of the motion and is not dynamically linked to the fermion operators a and $a^\dagger$ through Eq. (15).

We note that even though the zero-momentum limit of Eq. (22) exists and is nonzero, i.e.,

$$\lim_{k\to 0} \mathcal{V}_k^{\alpha\beta\gamma\delta}(\omega=0)=(V_0^{\alpha\beta})(V_0^{\gamma\delta})\left[\frac{\hbar d}{2\rho_4 c_{3s}}\right] \times \left[\frac{-2}{\hbar c_{3s}}\right](n_4^0 d) ,$$

this is not the effective potential of the tadpole or direct term of the Hartree-Fock energy. Conservation of momentum in the fermion loop of the tadpole diagram requires an identically zero momentum for the ripplon propagator and consequently vertices which are identically zero and which cannot be compensated for by the divergence of the ripplon propagator.

---

[1] M. D. Miller, Phys. Rev. B **17**, 1139 (1978); R. A. Guyer and M. D. Miller, *ibid.* **22**, 142 (1980).

[2] J. L. Cardy and D. J. Scalapino, Phys. Rev. B **19**, 1428 (1979).

[3] A. N. Berker and D. R. Nelson, Phys. Rev. B **19**, 2488 (1979).

[4] M. D. Miller and L. H. Nosanow, J. Low Temp. Phys. **32**, 145 (1978).

[5] J. P. Laheurte, J. C. Noiray, and J. P. Romagnan, J. Phys. (Paris) Lett. **42**, L-197 (1981).

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

[7] F. M. Ellis, R. B. Hallock, M. D. Miller, and R. A. Guyer, Phys. Rev. Lett. **46**, 1461 (1981).

[8] J. P. Laheurte, J. C. Noiray, J. P. Romagnan, and D. Sornette, J. Phys. (Paris) **47**, 39 (1986), and references cited therein.

[9] B. K. Bhattacharyya and F. M. Gasparini, Phys. Rev. B **31**, 2719 (1985); Phys. Rev. Lett. **49**, 919 (1982).

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

[11] D. J. Scalapino, in *Superconductivity,* edited by R. D. Parks (Marcel Dekker, New York, 1969), Chap. 10.

[12] L. M. Sander and H. B. Shore, Phys. Rev. B **11**, 4350 (1975).

[13] R. A. Guyer, M. D. Miller, and J. Yaple, Phys. Rev. B **25**, 4570 (1982).

[14] W. F. Saam and M. W. Cole, Phys. Rev. B **11**, 1086 (1975).

[15] J. Lekner, Philos. Mag. **22**, 669 (1970).

[16] For a discussion of ^{3}He surface states on the bulk liquid ^{4}He surface, see D. O. Edwards and W. F. Saam, in *Progress in Low Temperature Physics,* edited by D. F. Brewer (North-Holland, Amsterdam, 1978), Vol. 7A, p. 282.

[17]F. M. Gasparini, B. Bhattacharyya, and M. J. DiPirro, Phys. Rev. B **29**, 4921 (1984).

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

[19]D. S. Sherrill and D. O. Edwards, Phys. Rev. B **31**, 1338 (1985).

[20]*Handbook of Mathematical Functions,* edited by M. Abramowitz and I. A. Stegun (Dover, New York, 1965), Chap. 9.

[21]N. D. Mermin, Ann. Phys. (N.Y.) **21**, 99 (1963).

[22]J. des Cloizeaux, in *Many Body Problem,* Les Houches 1967, edited by C. deWitt and R. Balian (Gordon and Breach, New York, 1968), p. 1.

[23]See, for example, A. L. Fetter and J. D. Walecka, *Quantum Theory of Many-Particle Systems* (McGraw-Hill, New York, 1971).

[24]See Ref. 23, p. 401. Here $\int \zeta(\rho) d\rho = 0$ in order that the average film thickness be well determined.

[25]J. C. Owen, Phys. Rev. Lett. **47**, 586 (1981).

[26]E. M. Lifshitz and L. P. Pitaevskii, *Statistical Physics Part 2* (Pergamon, New York, 1980), Chap. III.

[27]See Ref. 16, and references cited therein.

[28]M. D. Miller, Phys. Rev. B **14**, 3937 (1976).

[29]X. Wang and F. M. Gasparini, Phys. Rev. B **38**, 11245 (1988).

[30]E. Krotscheck, Phys. Rev. B **32**, 5713 (1985).

[31]E. Krotscheck, M. Saarela, and J. L. Epstein, Phys. Rev. B **38**, 111 (1988).

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