culated density dependence (and hence the calculated pressure) to the location of the minimum of the potential, is potentially even more useful in that the second virial coefficient depends more on the shape of the bowl than on its location. However, we feel that this feature of the calculations will be useful only when more definitive calculations have been done, because the possibility that all the variational calculations (including the present one) have a wrong density dependence (even if the correct potential were used) cannot be ruled out. It is certainly true, for example, that different calculations, e.g., that of Massey and Woo[1] and the present one, both using the LJ potential, produce rather different density dependence, and we do not feel that the density dependence of the calculations done to date is sufficient to warrant any statement as to which of the potentials is most accurate.

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PHYSICAL REVIEW A VOLUME 5, NUMBER 1 JANUARY 1972

Growth of He^3 Layers from Surfaces of Nearly Saturated Solutions of He^3 in Superfluid He^4

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The problem of the growth of the He^3 phase from a free surface of a nearly saturated solution of He^3 in superfluid He^4 at zero temperature is considered. Using a Fermi-liquid theory modified to account for the finite thickness of the He^3 phase by introducing a distortion of the Fermi surface, we find that the number of He^3 layers N_L in the He^3 phase increases as the He^3 chemical potential μ_3 approaches its bulk value μ_3^0 as $N_L \sim (\mu_3^0 - \mu_3)^{-1/2}$. This prediction is in agreement with recent experimental results of Guo, Edwards, Sarwinski, and Tough.

I. INTRODUCTION

Here we consider the problem of the growth, at zero temperatures, of He^3 layers from a free, plane surface of a nearly saturated solution of He^3 in superfluid He^4. In order to understand the nature of this problem, it is useful to look qualitatively at what happens as He^3 atoms are added to a bath of superfluid He^4 which has a free, plane surface, and which initially contains no He^3. Both experimental[1-3] and theoretical[4] evidence indicate the existence of bound states for the He^3 atoms on the He^4 surface. As shown by Guo, Edwards, Sarwinski, and Tough,[3] as the He^3 atoms are added at 0 °K they first fill up the surface states until approximately one layer of He^3 is deposited on the He^4 surface, Subsequently, added He^3 atoms spill over into the bulk He^4, increasing the bulk He^3 concentration from zero toward its limiting value of 6.6% at the phase-separation curve. Now, the point on the phase-separation curve at $T = 0$ °K is the point at which the He^3 chemical potential μ_3 is equal to its value μ_3^0 in pure bulk He^3. As μ_3 approaches μ_3^0, then, some of the He^3 atoms being added begin to build a second layer of He^3 on the surface, followed by a third, and so on. A phase of pure He^3 grows on the surface of the solution, the number of layers N_L in this phase approaching infinity as $\mu_3 \to \mu_3^0$. And, as $N_L \to \infty$, X_3 in the substrate approaches 6.6%. There is no possibility of a metastable solution with $X_3 > 6.6\%$ being formed in the substrate since the initial He^3 layer on the surface acts as a nucleus for the growth of the He^3 phase.

We will use this picture, drawn by the experimentalists, as a starting point for our theory. Our goal is a somewhat limited one: We want to determine the asymptotic behavior of N_L as $\mu_3 \to \mu_3^0$. For very small N_L ($N_L \ll 1$) the zero temperature version of Andreev's noninteracting two-dimensional Fermi-gas theory[4] seems to be quite adequate for determining N_L as a function of μ_3.[3] For slightly larger N_L, Fermi-liquid corrections become important and could perhaps be discussed in terms of an effective-potential theory.[5] The treatment of small values of N_L (roughly $N_L \lesssim 10$) is not attempted here.

It seems reasonable to expect that the properties of a very thick surface He^3 film ($N_L \gg 1$) will be describable in terms of a quasiparticle picture not very different from that used for pure bulk He^3, and it is on this idea that our calculation is based. We argue that the dominant effect of the finite thickness of the He^3 film is to produce a slight distortion of the Fermi surface of bulk He^3. This distortion is characterized by introducing transverse (for motion parallel to the surface) and longitudinal effective masses differing from the bulk effective mass m_3^* by terms of order N_L^{-1}. This idea, coupled with the assumption that the important He^3 quasiparticle states in the system are, for large N_L, completely separable into bulk and surface states, leads to the result that $N_L \sim (\mu_3^0 - \mu_3)^{1/2}$ for μ_3 very close to μ_3^0. The proportionality constant is a measure of the distortion of the Fermi surface in the film. This asymptotic behavior for N_L is in agreement with recent experimental results of Guo, Edwards, Sarwinski, and Tough.[3]

II. THEORY

We commence the development of our theory with the assumption that a Fermi-liquid theory for the He^3 atoms is valid for all N_L. Such a theory involves, in principle, no more assumptions than those used in the theory of finite Fermi systems applied to nuclear matter.[6] Adding He^3 atoms to the system is then equivalent to adding quasiparticles to states (enumerated by index l) on the Fermi surface. Hence, if N_3 is the total number of He^3 atoms,

$$\frac{\partial N_3}{\partial \mu_3} = \sum_l \frac{\partial n_l}{\partial \mu_3} = \sum_l \frac{\partial n_l}{\partial \epsilon_l}\left(\frac{\partial \epsilon_l}{\partial \mu_3} - 1\right) , \tag{1}$$

using the usual expression for the quasiparticle distribution function

$$n_l = \Theta(\mu_3 - \epsilon_l) . \tag{2}$$

The ϵ_l are the quasiparticle energies, and the derivatives are taken at zero pressure, zero temperature, and constant surface area A. $\Theta(x)$ is defined by

$$\Theta(x) = \begin{cases} 1 & \text{for } x \geq 0 \\ 0 & \text{for } x < 0 . \end{cases}$$

Defining the density of states at the Fermi surface by

$$\nu(\mu_3) \equiv -\sum_l \frac{\partial n_l}{\partial \epsilon_l} = \sum_l \delta(\mu_3 - \epsilon_l) , \tag{3}$$

and writing

$$\frac{\partial \epsilon_l}{\partial \mu_3} = \frac{\partial \epsilon_l}{\partial N_3}\,\frac{\partial N_3}{\partial \mu_3} ,$$

we may easily put (1) in the form

$$\frac{\partial N_3}{\partial \mu_3} = \nu(\mu_3)\bigg/\left(1 - \sum_l \frac{\partial n_l}{\partial \epsilon_l}\,\frac{\partial \epsilon_l}{\partial N_3}\right) . \tag{4}$$

When N_L is very large, we assume that we may separate the set $\{l\}$ of quasiparticle states into one set of states associated with the pure He^3 phase and another associated with the He^3-He^4 mixture substrate. Then an equation like (4) holds separately for the number N_s of particles in the He^3 phase as well as for the number of He^3 atoms in the substrate. We then have

$$\frac{\partial N_s}{\partial \mu_3} = \nu_s(\mu_3)\bigg/\left(1 - \sum_l \frac{\partial n_l}{\partial \epsilon_l}\,\frac{\partial \epsilon_l}{\partial N_s}\right) . \tag{5}$$

Here, and in the following, the sum over states refers to states in the He^3 phase only. $\nu_s(\mu_3)$ is the density of these states at the Fermi surface. To specify N_L we define it in terms of the bulk He^3 number density n_{30} by

$$N_s \equiv A\, n_{30}^{2/3}\, N_L , \tag{6}$$

where A is the area. Then, in terms of N_L, a new density of states

$$\nu_L(\mu_3) \equiv \nu(\mu_3)/A\, n_{30}^{2/3} \tag{7}$$

and an average of $\partial\epsilon_l/\partial N_s$,

$$\nu_s(\mu_3)\left\langle \frac{\partial \epsilon_l}{\partial N_s}\right\rangle \equiv -\sum_l \frac{\partial n_l}{\partial \epsilon_l}\,\frac{\partial \epsilon_l}{\partial N_s} \tag{8}$$

over the Fermi surface, (5) may be put into the form

$$\frac{\partial N_l}{\partial \mu_3} = \nu_L(\mu_3)\bigg/\left(1 + \nu_L(\mu_3)\left\langle \frac{\partial \epsilon_l}{\partial N_l}\right\rangle\right) . \tag{9}$$

Note that in (9) the term $\nu_L(\mu_3)\langle \partial\epsilon_l/\partial N_l\rangle$ plays the role of a Fermi-liquid factor.

Before proceeding further, we introduce the thickness d of the surface He^3 film by

$$N_s \equiv A n_{30} d . \tag{10}$$

From (6), N_L and d are related by

$$N_L = n_{30}^{+1/3} d \ . \tag{11}$$

With these preliminaries out of the way, we can now introduce our model for a thick He^3 film. For a thick film, we expect the important states l to be rather like the quasiparticle states in bulk He^3, but that the effective mass m_T^* for motion parallel to the surface differs from that for motion perpendicular to the surface m_L^*, and that both differ by a term of order $1/N_L$ from the bulk He^3 effective mass m^*. We write

$$\begin{aligned} m_T^* &= m^* (1 + a_T/N_L) \ , \\ m_L^* &= m^* (1 + a_L/N_L) \ , \end{aligned} \tag{12}$$

where a_L and a_T are constants having order of magnitude 1. A quasiparticle state located near the Fermi surface will have energy

$$\epsilon = \epsilon_0 + \frac{p_L^2}{2m_L^*} + \frac{p_T^2}{2m_T^*} \ . \tag{13}$$

ϵ_0 is just the difference between μ_3 and the Fermi temperature,

$$\epsilon_0 = \mu_3 - k_B T_F \ , \tag{14}$$

and when $\vec{p}_L + \vec{p}_T$ is on the Fermi surface,

$$\left[\left(\frac{p_L^2}{2m_L^*}\right) + \left(\frac{p_T^2}{2m_T^*}\right)\right]_{\text{Fermi surface}} = k_B T_F \ . \tag{15}$$

The term ϵ_0 represents a sort of quasiparticle mean-field energy. We expect that its dependence on $d \sim N_L$ is determined predominately by potential energy effects, and that as a consequence it depends on N_L as

$$\epsilon_0 = \epsilon_{0B} (1 - b/N_L^3) \ , \tag{16}$$

where b is a constant and ϵ_{0B} is the bulk value for ϵ_0. The argument for this dependence is the one given by Landau and Lifshitz.[7] The effects of this N_L dependence of ϵ_0 are small compared to those due to our assumed N_L dependences of the effective masses [Eq. (12)], and we neglect them henceforth. The momenta p_L and p_T depend upon the size of the film according to

$$p_L = \frac{2\pi\hbar l}{d} = \frac{2\pi\hbar l}{N_L n_3^{-1/3}}, \quad p_T = \frac{2\pi\hbar l'}{\sqrt{A}} \ , \tag{17}$$

where l and l' are integers. Hence,

$$\frac{\partial p_T}{\partial N_L} = 0 \ , \quad \frac{\partial p_L}{\partial N_L} = -\frac{p_L}{N_L} \ . \tag{18}$$

Combining (12), (13), and (18), we obtain

$$\frac{\partial \epsilon}{\partial N_L} = -\frac{p_L^2}{m_L^* N_L} + \frac{p_L^2}{2m_L^{*2}} \frac{m^* a_L}{N_L^2} + \frac{p_T^2}{2m_T^{*2}} \frac{m^* a_T}{N_L^2} \ . \tag{19}$$

It then follows that (here $V = Ad$)

$$\sum_l \frac{\partial n_l}{\partial \epsilon_l} \frac{\partial \epsilon_l}{\partial N_l} = -\frac{2V}{(2\pi\hbar)^3} \int d^3p \, \delta\left(k_B T_F - \frac{p_T^2}{2m_T^*} - \frac{p_L^2}{2m_L^*}\right) \times \left(-\frac{p_L^2}{m_L^* N_L} + \frac{p_L^2}{2m_L^{*2}} \frac{m^* a_L}{N_L} + \frac{p_T^2}{2m_T^{*2}} \frac{m^* a_T}{N_L}\right) \ . \tag{20}$$

The integral may be done in a straightforward fashion, and we find

$$\sum_l \frac{\partial n_l}{\partial \epsilon_l} \frac{\partial \epsilon_l}{\partial N_L} = \frac{V (2m_T^*)(2m_L^*)^{1/2}}{\pi^2 \hbar^3} \frac{(k_B T_F)^{3/2}}{3N_L} \times \left(1 - \frac{m^* a_L}{2m_L^* N_L} - \frac{m^* a_T}{m_T^* N_L}\right) \ . \tag{21}$$

We now want to expand the effective mass terms in (21) in powers of $1/N_L$. Doing this and using $V = Ad = AN_L n_{30}^{-1/3}$, we obtain

$$\sum_l \frac{\partial n_l}{\partial \epsilon_l} \frac{\partial \epsilon_l}{\partial N_L} = \frac{A(2m^*)^{3/2}}{3\pi^2 n_{30}^{1/3} \hbar^3} (k_B T_F)^{3/2} \left(1 - \frac{C}{N_L^2}\right) \ , \tag{22}$$

where, for brevity, we have written

$$C = \tfrac{1}{2} a_L (a_T - \tfrac{1}{4} a_L) \ . \tag{23}$$

We must now treat the term $k_B T_F$. Since, to order $1/N_L^2$, $\epsilon_0 = \epsilon_{0B}$, we have

$$\mu_3^0 - \mu_3 = k_B T_F^0 - k_B T_F + O(1/N_L^3) \ . \tag{24}$$

μ_3^0 and T_F^0 are the bulk He^3 chemical potential and Fermi temperature. Hence,

$$\begin{aligned} (k_B T_F)^{3/2} &= (k_B T_F^0)^{3/2} \left(1 - \frac{(\mu_3^0 - \mu_3)}{k_B T_F^0}\right)^{3/2} \\ &\approx (k_B T_F^0)^{3/2} \left(1 - \frac{3(\mu_3^0 - \mu_3)}{2 k_B T_F^0} + \cdots\right) \ . \end{aligned} \tag{25}$$

Further, since

$$k_B T_F^0 = \hbar^2 (3\pi^2 n_{30})^{2/3} / 2m^* \ , \tag{26}$$

we may combine (22)–(26) to find

$$\sum_l \frac{\partial n_l}{\partial \epsilon_l} \frac{\partial \epsilon_l}{\partial N_L} = A n_{30}^{2/3} \left(1 - \frac{C}{N_L^2} - \frac{3(\mu_3^0 - \mu_3)}{2 k_B T_F^0}\right) \tag{27}$$

for large N_L.

The density of states $\nu_L(\mu_3)$ is easily determined to be

$$\begin{aligned} \nu_L(\mu_3) &= \frac{m^* p_F^0}{n_{30} \pi^2 \hbar^3} N_L \left[1 + O\left(\frac{1}{N_L}\right)\right] \\ &= \frac{3}{2k_B T_F^0} N_L \left[1 + O\left(\frac{1}{N_L}\right)\right]. \end{aligned} \tag{28}$$

Putting (27) and (28) in (9) and noting that

$$\nu_L(\mu_3) \left\langle \frac{\partial \epsilon_l}{\partial N_L} \right\rangle \equiv \frac{-1}{A n_0^{2/3}} \sum_l \frac{\partial n_l}{\partial \epsilon_l} \frac{\partial \epsilon_l}{\partial N_L} \ , \tag{29}$$

we arrive at

$$\frac{\partial N_L}{\partial \mu_3} = \frac{3N_L}{2k_B T_F^0} \Bigg/ \left(\frac{C}{N_L^2} + \frac{3(\mu_3^0 - \mu_3)}{2k_B T_F^0} \right) , \tag{30}$$

keeping only the most divergent terms as $N_L \to \infty$. It is easily verified that (30) has the solution

$$N_L = \left(\frac{3k_B T_F^0 C}{2(\mu_3^0 - \mu_3)} \right)^{1/2} . \tag{31}$$

Equation (31) is the result at which we were aiming. The parameter C [see Eq. (23)] is a measure of the deformation of the Fermi surface due to the finite thickness d. The square-root divergence is thus, in our model, a purely quantum effect. Had we ignored the quantum character of the problem, ϵ_0 would have served as our chemical potential, and we would have gotten, directly from (16), a cube-root divergence as do Landau and Lifshitz.[7]

III. DISCUSSION

The form of the result (31) is in agreement with the experimental results of Guo, Edwards, Sarwinski, and Tough[3] when the constant C has the approximate value

$$C = \tfrac{1}{2} a_L (a_T - \tfrac{1}{4} a_L) \approx 0.3 . \tag{32}$$

A rough check on the consistency of our theory can be made by looking at the entropy per particle S_s of the He^3 phase (the "surface" entropy). In our theory, S_s differs from the value S_{30} for bulk He^3 due to the Fermi-surface distortion. We easily find

$$S_s = S_{30} \left[1 + \frac{a_T + \frac{1}{2} a_L}{N_L} + O\left(\frac{1}{N_L^2} \right) \right] . \tag{33}$$

Experiment[3] confirms the form of Eq. (33). It is, however, not very meaningful to attempt to extract a value for the coefficient $a_T + \frac{1}{2} a_L$ from the experimental data since our theory treats the surfaces of the He^3 film in a very crude fashion. The details of these surfaces will certainly be important in determining the coefficient of the $1/N_L$ term in the brackets in (33).

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Criticism of the Single-Particle-Green's-Function Approach to the Excitation Spectrum of Superfluid Liquid Helium*

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The experimentally observed spectrum for superfluid liquid helium has properties which are characteristic of a sound spectrum but in disagreement with the predictions of a naive use of the single-particle-Green's-function approach. It is argued that the dynamical origins of the spectrum are closely related to those which give rise to the sound propagation in normal liquids, and therefore that the helium spectrum should be sought in structures and formalisms that would be used to study sound in normal liquids.

I. THEORY VS EXPERIMENT

The condensate density is not a parameter which we can vary independently, but we can judge its relevance to the helium spectrum indirectly by considering the effects of increasing density or temperature. In both cases the condensate density may be presumed to decrease, since the kinetic energy of the liquid is increasing. The relevant observations are (a) The spectrum measured at saturated vapor pressure by neutron scattering[1,2] shows very little shift (but considerable broaden-