PHYSICAL REVIEW B VOLUME 59, NUMBER 5 1 FEBRUARY 1999-I

Excitations in thin ^{3}He-^{4}He superfluid films

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(Received 5 October 1998)

We explore the behavior of ^{3}He single-particle states in thin ^{4}He superfluid films by analyzing both third sound and magnetization data. The third-sound speed is sensitive to the transverse, quantum-mechanical extent of motion of the adsorbed ^{3}He. We are thus able to extract *from experiment* the size of the ^{3}He surface ground state. As this motion is state-dependent, we can interpret a high coverage feature as the signal that the first excited state has begun to be populated. For a 13.2 Å ^{4}He film on Nuclepore, we find that the surface state is $\approx$3.9 Å thick and that the onset coverage for first excited-state occupation is $\simeq$0.6 (i.e., $\simeq$0.039 Å^{-2}). This result combined with an analysis of magnetization step data show that the onset coverage *decreases* with increasing film thickness in the thin-film limit. [S0163-1829(99)01105-4]

In recent years, there has been considerable effort in trying to understand the properties of ^{3}He-^{4}He mixture films.[1] In previous work on (unsaturated) mixture films, it was shown that the presence of ^{3}He (Ref. 2) or other impurities[3] in the superfluid film alters the speed of third sound. The change in speed is a function of both the amounts of ^{3}He and also its physical location in the film. In thin mixture film systems such as those studied recently by Hallock and co-workers[1] the ^{3}He component occupies a well-defined set of single-particle levels (transverse, particle-in-a-box types of states[4]). A third sound measurement can detect the onset of filling of the excited states as a function of ^{3}He coverage because of orthogonality which locates each ^{3}He eigenstate slightly differently within the ^{4}He film.

Much of the recent interest in these mixture films was stimulated by the work of Bhattacharyya and Gasparini.[5] In a series of heat-capacity studies, they observed the presence of the ^{3}He transverse excited states[6,7] and also found a feature in the specific heats which was interpreted as a possible signal of condensation in the ^{3}He subsystem. In previous theoretical work,[8,9] a simple model was introduced in which the possible condensation would be driven by the ^{3}He-^{3}He effective interaction due to the exchange of a film excitation (the one ripplon exchange potential, OREP). This model was unable to account for condensation, but it did provide a mechanism for ^{3}He promotion into the first transverse excited state at an areal density well below one monolayer coverage. This result was surprising because from naive Fermi-gas arguments one would presume that the level spacing from the ground state to the first excited state is simply too wide to be crossed by a filled Fermi sea at submonolayer densities. The occupation of transverse excited states in a submonolayer ^{3}He system is directly observed in the magnetization steps observed by Hallock and co-workers.[10,11] In this paper, we shall point out that the occupation of the transverse excited states is also directly seen in the third-sound experiments of Sheldon and Hallock.[12] Further, we shall show that analysis of the third-sound data yields a measurement of the thickness of the ^{3}He surface ground state. Finally, by combining an analysis of the magnetization steps data and the third-sound data we can extract the onset ^{3}He coverage for first excited-state occupation as a function of ^{4}He film thickness.

We first examine third sound in a mixture film system with the ^{3}He component in its transverse ground state, i.e., the low ^{3}He-coverage limit.[13] In Refs. 2 and 3, it was shown that the expression for third sound in a ^{3}He-^{4}He mixture film is given by

$$\left(\frac{c_3^2}{c_{30}^2}\right)=1-\frac{\rho_u}{\rho_l}\left[\frac{f_u(h_l)-f_u(h_l+h_u)}{f_l(h_4)}\right], \tag{1}$$

where the quantity $c_{30}^2=-(h_4-h_0)f_l(h_4)$ is the third-sound speed for pure ^{4}He. The l and u subscripts correspond to a two-layer model of the mixture. In the low-temperature limit, the ^{3}He is confined to the upper film of thickness h_u and the lower mobile layer, l, consists of the superfluid ^{4}He. In Eq. (1) the ρ's are mass densities, $h_l=h_4$ is the height of the mobile film above the substrate, and h_0 is the thickness of the immobile ^{4}He layer next to the substrate. In the simplest cases, the force per unit mass due to the van der Waals interactions is $f_x(h)=-3\alpha_s/(m_xh^4)$, where α_s is the substrate-helium van der Waals parameter and the m_x are film averaged masses. It is convenient to present the third-sound results in the form of Eq. (1) because then the often imprecisely known van der Waals parameters cancel out.

For almost all of the data reported in Ref. 2, the ^{3}He upper films were greater than a monolayer and so adding ^{3}He to the system simply increased h_u in a continuous manner. For a continuous growth model, we can replace the mass

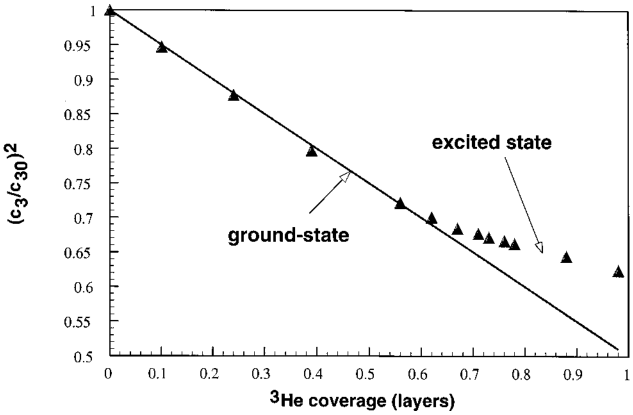

FIG. 1. Third sound squared as a function of ^{3}He coverage. The triangles are the data of Sheldon and Hallock (Ref. 12) and the line is the ground-state theory of Eq. (3). The change in slope at a coverage of 0.6 is the onset of excited-state occupation.

densities by their bulk equivalents, $\rho_u = m_3 n_3^0$ and $\rho_l = m_4 n_4^0$, so that Eq. (1) can be written

$$\left(\frac{c_3^2}{c_{30}^2}\right)_{\text{cont growth}} = 1 - \frac{n_3^0}{n_4^0}\left[1 - \left(1 + \frac{h_u}{h_4}\right)^{-4}\right], \tag{2}$$

where n_3^0 and n_4^0 are the bulk ^{3}He and ^{4}He number densities, respectively. In this model, all of the information concerning changes in the third-sound speed due to the addition of ^{3}He to the system is contained in the magnitude of h_u. For submonolayer ^{3}He, this model may not be sensible. Adding ^{3}He atoms in that case changes the areal density in the surface ground state but does not affect the ''thickness'' of the film, a quantity which is fixed by the transverse extent of motion of the adsorbed ^{3}He atom. If we try and take account of the quantum-mechanical transverse motion of the ^{3}He atom from the beginning then we are led to a picture different from a classical continuous growth model. We can imagine a ''box'' of area A and thickness h_u. As ^{3}He is added to this ''box'' the only effect at low densities is to change the ratio of the mass density in the upper film to that of the lower film.

Thus, in this picture, Eq. (1), immediately reduces to

$$\left(\frac{c_3^2}{c_{30}^2}\right)_0 = 1 - \Delta_{l0}\theta_3, \tag{3}$$

where $\theta_3 = (\sigma_3/\sigma_{3\max})$ is the coverage in units of monolayers, $\sigma_{3\max} = (n_3^0)^{2/3} \approx 0.065$ Å^{-2} is the areal density at monolayer completion, and the slope is given by

$$\Delta_{l0} = \frac{n_3^0}{n_4^0}\left[1 - \left(1 + \frac{h_u}{h_4}\right)^{-4}\right]. \tag{4}$$

In the low coverage regime where the ^{3}He only occupies the transverse ground state, c_3^2/c_{30}^2 should be a linearly decreasing function of the coverage with a slope given by Eq. (4). We note that the slope Δ_{l0} depends only on the thickness of the superfluid film and is *independent of the substrate*.

In Fig. 1 we compare Eq. (3) with the experimental data of Sheldon and Hallock.[12] In this system, $h_4 = 3.67 l_4$, and $l_4 = (n_4^0)^{1/3} = 3.6$ Å, is the thickness of one ^{4}He monolayer. Excellent agreement with the experimental data is obtained by fixing the parameter $h_u = 3.9$ Å . The parameter h_u is a measure of the thickness of the ^{3}He transverse ground-state probability density and we note in passing that this particular value for h_u is the conventional ^{3}He layer thickness l_3, where $l_3 = (n_3^0)^{1/3} = 3.9$ Å. Thus, with $h_u = l_3$ we find $\Delta_{l0} = 0.5$, which is in excellent agreement with the experimental data. We note that the continuous growth model also shows a linear decrease in c_3^2 with increasing coverage in the low coverage limit. In that case the slope is given by $\Delta_{\text{cont growth}} = (4\sigma_{3_{\max}}/n_4^0 h_4)$, and for the system of Ref. 12, $\Delta_{\text{cont growth}} \simeq 0.9$, nearly a *factor of two* in disagreement with experiment.

The third-sound experimental data in Fig. 1 shows a change in slope at a coverage ≈ 0.6. We interpret this change in slope as the signal of the onset of occupation of the first excited state. The fact that the slope bends up in Fig. 1, that is, the third-sound speed increases relative to what the third-sound speed would be if all the ^{3}He were in the ground state, is an important constraint on models for third sound in this region. A second constraint, as will be discussed further on, comes from an analysis of the magnetization step data of Ref. 10.

There have been a number of calculations of the wave functions for the ^{3}He transverse excited states.[4,8,14] The first excited-state wave function has a larger transverse extent than the ground state, which we interpret as a larger value for the parameter h_u. The change in the value of h_u affects the third-sound speed in two ways. First, by increasing the thickness of the normal fluid layer, the third-sound speed is decreased. Second, by decreasing the mass density in the normal fluid layer, the third-sound speed increases. For agreement with experiment, the latter effect must dominate the former.

Let the upper film thickness $h_u = l_3 + \Delta h_{31}$ where Δh_{31} is a single adjustable parameter fixed by the third sound data of Fig. 1. Then, in the spirit of this model, the third-sound speed in the regime $\sigma_3 > \sigma_{\text{on}}$, where σ_{on} is the onset density for first excited-state occupation, can be written

$$\left(\frac{c_3^2}{c_{30}^2}\right)_1 = \left(\frac{c_3^2}{c_{30}^2}\right)_{\text{on}} - \Delta_{l1}(\theta_3 - \theta_{\text{on}}), \tag{5}$$

where Δ_{l1} is given by Eq. (4) divided by h_u/l_3. If we define

$$\Delta\left(\frac{c_3^2}{c_{30}^2}\right) = \left(\frac{c_3^2}{c_{30}^2}\right)_1 - \left(\frac{c_3^2}{c_{30}^2}\right)_0$$

then from Eq. (5)

$$\Delta\left(\frac{c_3^2}{c_{30}^2}\right) = (\Delta_{l0} - \Delta_{l1})(\theta_3 - \theta_{\text{on}}). \tag{6}$$

In order to be in agreement with the data of Fig. 1, we need to require that the difference in slopes $\Delta_{l0} - \Delta_{l1} \approx 0.3$. This requires $\Delta h_{31} = 2.8 l_3$. This rather large value for Δh_{31} is indicative of the surprisingly large change in slope caused by excited-state occupation. A better understanding of the third-sound speed in this coverage regime will require a better theoretical picture of the ^{3}He probability distribution in the excited state.

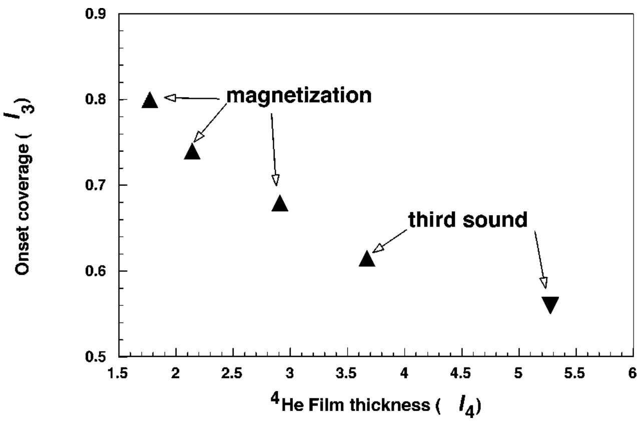

FIG. 2. Onset coverage for first-excited state occupation as a function of ^{4}He film thickness. The three low coverage points are from the analysis of the magnetization data as discussed in the text. The point at $3.67 l_4$ is taken from Fig. 1. The upside-down triangle at $5.274 l_4$ is perhaps a lower limit determined by an unpublished third-sound run.

A reanalysis of the magnetization data of Refs. 10 and 11 also gives important *new* information for the mixture film system. The areal density at the beginning and end of the first magnetization plateau is sufficient to determine the onset coverage of first excited-state occupation. Define σ_l (σ_r) to be the density at the left (right) corner of the plateau. Then, $\sigma_{\rm on}=\sigma_l+\sigma_r$. Following this, the level spacing between the energies of the transverse ground state and first excited state is $\Delta\epsilon=\hbar^2\pi\sigma_{\rm on}/m_3^*$, and the fractional population in the first excited state at monolayer completion is $x_1(1)=\frac{1}{2}(1-\theta_{\rm on})$. We analyze the results of measurements made at three ^{4}He film thicknesses: $1.77 l_4$, $2.14 l_4$, and $2.91 l_4$.

We find for the fractional population of the first excited state at monolayer completion $x_1(1)=0.10$, 0.13, and 0.16, respectively. These small values are an important constraint on viable third-sound models for the excited state. They seem to rule out continuous growth-type models in which one assumes that the main affect of promoting particles into the first excited state is to increase the value of h_u in a continuous manner (i.e., by adding the density of particles in the first excited state uniformly to the ground-state box $h_u=l_3$). These models yield values of $x_1(1)\approx 0.5$ after requiring them to be in agreement with the third-sound data of Fig. 1. In addition, these results also seem to rule out ''independent-gas'' types of models. That is, models which assume that the ground-state and first excited-state populations make independent additive contributions to Eq. (1). Once more they seem to need far more atoms in the excited state to yield agreement with the third-sound data than is permitted by the magnetization data.

In Fig. 2 we plot $\sigma_{\rm on}$ obtained from the three magnetization experiments together with the value obtained from Fig. 1, for the third-sound analysis at $3.67 l_4$. The fit is quite consistent. The decrease in the onset coverage as a function of increasing ^{4}He film thickness is due to the increase in the level spacing, $\Delta\epsilon$, with *decreasing* film thickness. The above analysis yields $\Delta\epsilon\approx 1.9$, 1.8, 1.7 K for the three films, respectively. (For these results we use $m_3^*=1.38 m_3$ the hydrodynamic effective mass.) These spacings are in agreement with the measured values of Alikacem, Sprague, and Hallock.[15] The increase in the level spacing is a thin-film phenomenon due to the proximity of the free surface to the well of the substrate potential. This can be seen in the calculations of Refs. 4,8. We note that there is also both experimental, Ref. 7, and theoretical, Ref. 9, evidence that $\Delta\epsilon$ is a function of θ_3 in addition to h_4.

The data point at $h_4=5.274 l_4$ in Fig. 2 corresponds to the highest coverage reached in an unpublished third-sound run. The data shows a very slight upward curvature for coverages out to $\leqslant 0.56 l_3$. If this is the linear, low coverage ground-state occupation region then the onset coverage for first excited-state occupation is at coverages greater than $0.56 l_3$. The slope of this third-sound data is close to 0.5 rather than the 0.4 obtained from Eq. (4). If h_u, in Eq. (4) is a function of h_4 then one can fit this data by using $h_u\approx 1.44 l_3$. We note, from Eq. (4), that in the thick-film limit the slope Δ_{l0} vanishes like h_4^{-1}. More data is needed to investigate the dependence of both $\theta_{\rm on}$ and Δ_{l0} on h_4.

In Refs. 8 and 9 a semiphenomenological model for a mixture film system was introduced in order to carefully examine the possible role played by the ^{3}He-^{3}He effective interaction due to exchange of a ripplon (OREP). In previous calculations, the spectrum corresponding to Eq. (2) was used. As shown above, however, the appropriate spectrum is that of Eq. (3) which is smaller by almost a factor of 2. Thus, we are led to repeat our calculations with the proper film excitation spectrum. We note also that here we shall use the value $\alpha_s=1900$ K$-$Å^3 for the Nuclepore-helium van der Waals parameter as reported by Higley, Sprague, and Hallock.[10] It is slightly larger than the value 1740 K$-$Å^3 [4] which was previously used. We find that $\theta_{\rm on}=0.6$ and that $x_1(1)=0.08$. The onset coverage is in nearly exact agreement with the third-sound results shown in Fig 1. The excited-state population is approximately a factor of 2 smaller than the results estimated from the analysis of the magnetization data discussed above. We note however that a recently introduced sophisticated time-dependent variational method may offer an alternative *single-particle* view of the ^{3}He excited-state structure as discussed above.[16]

Finally we point out that, in principle, heat-capacity experiments on this system can directly verify the above picture concerning the onset of excited-state occupation as kinks in the third-sound signal. At a fixed temperature, small relative to the Fermi energy at onset (≈ 1.8 K), the two-dimensional heat capacity is simply proportional to the number of occupied states (assuming a state independent effective mass). Thus, at fixed *temperature* the heat capacity, as a function of ^{3}He coverage, will exhibit a steplike structure with the steps occurring as the Fermi energy reaches each new excited state.[17,18] The step structure becomes rounded at higher temperatures, however, it ought to be straightforward to see the predicted step at $\theta_{\rm on}=0.6$ for the $3.67 l_4$ system studied by Sheldon and Hallock.[12]

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