Third sound and energetics in ^{3}He-^{4}He mixture films

P. A. Sheldon and R. B. Hallock
Laboratory for Low Temperature Physics, Department of Physics and Astronomy, University of Massachusetts at Amherst, Amherst, Massachusetts 01003
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We report measurements of the third-sound frequency and attenuation in a capacitive resonator as a function of temperature for various submonolayer densities of ^{3}He ($0.007 \le n_3 \le 0.049$ Å^{-2}) in a thin ^{4}He film ($n_4 = 0.389$ Å^{-2}), which result in a determination of the ground-state and first-excited-state energies for the ^{3}He which are in good agreement with the energetics determined by NMR. Interesting features are seen in the temperature dependence of the frequency and phase of the third sound for $100 < T < 200$ mK.

Helium mixture films bound to a substrate are rich with information about the quasi-two-dimensional properties of Fermi systems. In a ^{4}He film ^{3}He occupies bound states perpendicular to and free-particle states parallel to the substrate.[1] In its lowest energy state, a ^{3}He atom resides at the surface of the ^{4}He film. The ^{3}He has higher energy states available to it, ε_i, which are apparently located inside the ^{4}He film. Bhattacharyya, Di Pirro, and Gasparini[2] measured the bound-state energies of ^{3}He on a ^{4}He film using heat-capacity techniques. More recently the ground-state energy, ε_0, and the energy difference between the ground state and first excited state, Δ, has been measured[3,4] using NMR for a large range of ^{4}He film thicknesses with a fixed coverage of ^{3}He, and for many submonolayer coverages of ^{3}He on a fixed thickness of ^{4}He.[5] We report here measurements of the temperature and ^{3}He density dependence of the frequency, damping and phase of third-sound resonance modes. The energetics of the ^{3}He Fermi system are extracted from the data and compared to our results for the energetics determined[5] by NMR techniques. We document unexplained behavior: a weak but persistent maximum in the third-sound frequency near 150 mK and a dramatic temperature dependence of the phase for $150 < T < 200$ mK.

Third sound[6] is a temperature and areal density wave which propagates on superfluid helium films. Such films can exist on most solid surfaces due to the van der Waals attractive interaction between the helium atoms and the atoms of the solid substrate. The third-sound velocity for a pure ^{4}He film is given by the approximate expression[7]

$$C_{30}^2 \approx \rho_s/\rho(1-D/d_4)\vartheta d_4[1+TS/L]^2 ,$$

where ρ_s/ρ is the bulk superfluid fraction, D is the thickness of the immobile part of the film adjacent to the substrate, d_4 is the thickness of the film, and ϑ, S, L are the van der Waals force, entropy, and latent heat per unit mass. Over the temperature range of this work, $TS/L \simeq 0$, $\rho_s/\rho \simeq 1$, D is constant,[8] and ϑ varies only with ^{4}He film thickness.

The third-sound frequency (f), quality factor (Q), amplitude (A) and phase between the drive and detected amplitude (Φ) are measured with a third-sound resonator which is mounted in a copper cell alongside an NMR coil. The cell is attached to a dilution refrigerator. The third-sound resonator is constructed from a rectangular piece of Nuclepore, 14.3×19.1 mm. The Nuclepore is 10 μm thick, with $\sim 3 \times 10^8$ pores/cm^2, and each pore has a 2000 Å diameter. 90% of the surface area is inside the pores. A silver strip is evaporated onto one end of the resonator to drive the third sound thermally, and capacitor plates are evaporated onto both sides at the opposite end to detect thickness changes capacitively. The capacitor is part of an LC tank circuit driven by a tunnel diode oscillator which oscillates at a nominal frequency of 25 MHz. When the third-sound modes are driven (~ 300 Hz) the film thickness between the capacitor plates changes sinusoidally. The resulting frequency modulation is detected with a phase lock loop, and the amplitude of a driven frequency is measured by lock-in detection with the drive frequency as reference. The sensitivity of the frequency of the LC circuit to film thickness is measured as 30 Hz/Å; we are able to measure third-sound film thickness changes of 2.5 μÅ. f, A, Q, and Φ for a third-sound mode are measured by sweeping the drive through the resonant frequency, and performing a four-parameter fit of the in-phase and quadrature amplitudes versus frequency to a Lorentzian. All data reported here are for the fundamental mode f_1.

We vary the amount of ^{3}He in a film of ^{4}He, $n_4 = 0.389$ Å^{-2} ($d_4 = 3.65$ layers). For each of the eight ^{3}He densities in the range $0.007 \le n_3 \le 0.049$ Å^{-2} ($0.099 \le d_3 \le 0.721$ layers) described here, we made measurements in the temperature range $40 \le T \le 500$ mK for drive powers $0.044 \le P_{dr} \le 440$ nW. As expected, the amplitude goes to zero at zero drive power, and it rises linearly with drive power for small P_{dr}.[9] The frequency is observed to be a linear function of drive power, and can be extrapolated back to zero drive power with negligible error. The Q is independent of drive at the lower drive powers, except apparently at the very lowest drive powers where it is also quite imprecise. Unless otherwise specified, the frequency reported here is extrapolated to $P_{dr} = 0$. The Q's at the lowest three drive powers are extrapolated to obtain the $P_{dr} \to 0$ limit.

A simple quantitative description for the third-sound velocity in a mixture film system of ^{3}He and ^{4}He can be derived from a set of linearized hydrodynamic equations[10,11] by assuming that there are two layers; the lower

layer (l) contains all the superfluid and some concentration of ^{3}He and the upper layer (u) is a normal fluid blanket of ^{3}He. The theory assumes that the helium is incompressible and that there are no interaction effects between the two layers. The result for the third-sound velocity in a mixture film at $T=0$ is

$$C_3^2=C_{30}^2\left[\frac{m_4}{m_l}\right]\left[\frac{d_4}{d_l}\right]^4\left[1-\frac{\rho_u}{\rho_l}+\frac{\rho_u/\rho_l}{(1+d_u/d_l)^4}\right], \quad (1)$$

where C_{30} is the third-sound velocity for a film of pure ^{4}He of thickness d_4, and d_4 is the thickness of the film if there were no ^{3}He, m_4 is the mass of a ^{4}He atom, m_l is the average mass per particle in the lower layer, ρ_u and ρ_l are the average number densities in the upper and lower layers, and d_u and d_l are the thicknesses of those layers. We can add temperature dependence to the film thicknesses d_u, d_l in Eq. (1) by assuming that as the temperature increases, the ^{3}He is both excited into its higher-energy state inside the ^{4}He film and evaporated into the vapor. Thus the various parameters of Eq. (1) can be expressed in temperature-dependent forms which depend on the energetics of the ^{3}He.

In order to calculate the ^{3}He population densities of the bound states and the vapor, we need to know the chemical potential everywhere. The chemical potential of a dilute vapor of ^{3}He atoms is $\mu_v=k_BT\ln(1/2\rho_v\lambda_T^3)$, where ρ_v is the ^{3}He number density in vapor and $\lambda_T=\sqrt{2\pi\hbar^2/mk_BT}$ is the thermal de Broglie wavelength. Edwards *et al.*[12] propose a simple interaction term in the chemical potential of the film, $\mu_f=\varepsilon_B+1/2n_3V_0^S+\mu_{\rm IFG}$, where n_3 is the ^{3}He areal density, ε_B is the ^{3}He binding energy to the film, V_0^S is a quasiparticle interaction potential and $\mu_{\rm IFG}=k_BT\ln[\exp(n_3\pi\hbar^2/m^*k_BT)-1]$ is the chemical potential of an ideal two-dimensional Fermi gas. The effective mass m^* of a ^{3}He atom in a mixture film is derived at low ^{3}He densities using the susceptibility measurements[13] from NMR and Fermi-liquid parameters calculated by Krotscheck, Saarela, and Epstein.[14,15] We measure $m^*=1.5m$, and use this value for the higher ^{3}He coverages since it is predicted[15,16] to change very little over our range of densities. Bhattacharyya, DiPirro, and Gasparini[2] also show that there is little ^{3}He dependence of m^* above $n_3\approx0.01$ Å^{-2}. Equating the expressions for μ_v and μ_f, the fraction of ^{3}He atoms which evaporate from the film, $p_v\equiv\rho_vV/N$, where V is the volume of the sample cell, can be determined if we add the additional constraint that particle number is conserved, $N=\rho_vV+n_3A$, where A is the area of the film.

Sprague *et al.*[3] were able to solve this problem analytically by making a number of reasonable assumptions. In calculating the number of ^{3}He atoms in the vapor they found that they did not need to include the effects of evaporation into the first excited state because it has an energy very near to the vapor phase, and has a phase space which is smaller by a factor of V/λ_TA. At temperatures where evaporation is prevalent (above 300 mK), there is very little ^{3}He mixed into the lower ^{4}He layer, and the ^{3}He in the vapor will dominate the effect of any ^{3}He in an excited state. For our higher ^{3}He densities the quasiparticle interaction term is small but not negligible; using the value for ^{3}He on bulk ^{4}He, $V_0^S=11.6$ K Å^2,[17] $\frac{1}{2}n_3V_0^S$ is 5% of ε_B for our highest density. We use μ_f exactly, and still ignore the negligible effects of ^{3}He evaporating into higher-energy states in the film at temperatures above 300 mK. This results in a nonanalytic function for the fraction of ^{3}He atoms left in the film during evaporation, and that quantity can be determined numerically.

For the ^{3}He in the film, we need to derive the fraction of ^{3}He atoms in each state, n_iA/N, at all temperatures. For lower ^{3}He densities it is adequate to use a Boltzmann distribution for a two-state system so that $n_1A/N=\exp(-\Delta/k_BT)$, where $\Delta+\epsilon_F=\varepsilon_1-\varepsilon_0$. For higher densities, the film has the properties of a two-dimensional Fermi gas. We use the chemical potential for each energy state in the film, $\mu_i=\varepsilon_i+\frac{1}{2}n_iV_0^S+\mu_{{\rm IFG}_i}$, where ε_i, n_i, and $\mu_{{\rm IFG}_i}$ are the energy, areal density, and ideal Fermi gas chemical potential of state i, respectively. Conserving the number of ^{3}He atoms, $N=\sum_iN_i$, and noting that the chemical potential is the same everywhere, we can solve for the ground- and first excited states to give a nonanalytic equation for one quantity, either $p_0\equiv n_0A/N$ or $p_1\equiv n_1A/N$, which can be solved numerically. With the numerical functions for the probability distributions p_v, p_0, and p_1 we interpret the features seen in the third-sound data and extract values for the bound-state energies, comparing them to those found from NMR.

The temperature dependence of the third-sound frequency is shown in Fig. 1(a) for eight different ^{3}He densities. As seen previously,[11] at low temperatures the third-sound frequency has the expected dependence on ^{3}He thickness according to Eq. (1) for a phase-separated film (the lower layer being *nearly* all ^{4}He and the upper, ^{3}He). The film is expected to be completely phase separated at $T=0$ as determined by the energetics because the ground state of the ^{3}He is at the surface of the ^{4}He film.[1]

At low temperatures ($T\le150$ mK) the ^{3}He is in its ground state at the surface of the ^{4}He film, and the frequency has little structure. There is a small maximum at 150 mK which gets relatively larger as ^{3}He density is increased. This is similar to the anomaly reported earlier.[18] This peak was seen to be much more dramatic[19] at considerably higher ^{3}He densities. The frequency decrease in the range 150 mK$\le T\le$300 mK is attributed[20] to the ^{3}He being excited into a higher-energy state. If the higher-energy state is inside the ^{4}He film, then the film becomes effectively thicker, and the third-sound frequency should decrease. For the higher ^{3}He densities reported here, the application of Eq. (1) with the temperature dependence included in the layer thicknesses leads us to expect a more gradual decrease in frequency with increased temperature for any reasonable energy difference Δ between the first two states than is observed. For $T>300$ mK the frequency increases due to ^{3}He evaporating from the film. We can fit Eq. (1) to the increasing part of the frequency vs temperature curve, with the binding energy in the probability density of the vapor as a parameter. In the region where evaporation is prevalent, as argued above, the ^{3}He evaporating from the top of the ^{4}He dominates the effect of the ^{3}He mixing into the ^{4}He

film, so we use the phase-separated form of Eq. (1),

$$\left[\frac{C_3(T)}{C_{30}(T)}\right]^2 = 1 - \frac{\rho_3}{\rho_4}\left[1 - \left[1 + \frac{d_3(T)}{d_l}\right]^{-4}\right], \qquad (2)$$

where d_l is the lower layer thickness needed to account for the frequency reduction between the lowest temperature (where it is truly a layered film) and the minimum of the f vs T curve, presumably due to a small amount of ^{3}He mixed into the lower layer. From the fits of Eq. (2) to data, we can determine the ground-state binding energy of ^{3}He to the ^{4}He film for each ^{3}He density. The fits to the evaporation region are shown in Fig. 1(a) for selected ^{3}He densities as smooth curves.

The excess attenuation is the additional attenuation as ^{3}He is added to the ^{4}He film, $R_{ex} \equiv 1/Q - 1/Q_0$, where Q_0^{-1} is the attenuation for the lowest ^{3}He density, $n_3 = 0.007$ Å^{-2}. The excess attenuation R_{ex} is observed to be thermally activated[20,21] with an empirical form $R_{ex} = R_0 + R_1 p_1$, where R_0 and R_1 are arbitrary parameters and p_1 is the first-excited-state probability distribution for a two-state system with activation energy Δ'.

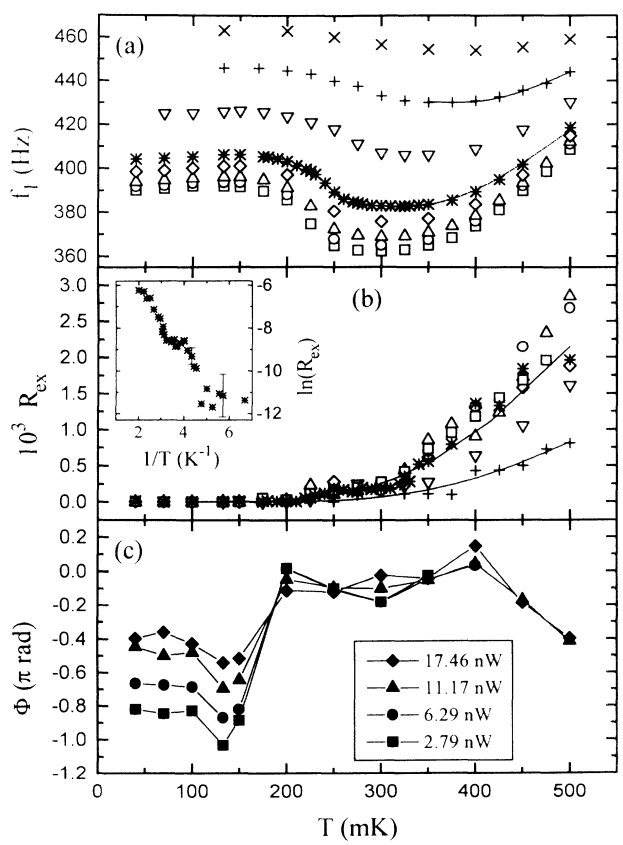

FIG. 1. Temperature dependence of (a) f_1 and (b) R_{ex} ($R_{ex} = 1/Q - 1/Q_0$, where $1/Q_0$ is the damping for the lowest ^{3}He coverage) for each ^{3}He coverage: $n_3 = 0.007$ (×), 0.016 (+), 0.025 (▽), 0.036 (∗), 0.040 (◇), 0.044 (△), 0.046 (○), and 0.047 Å (□). Typical fits are shown to the frequency and damping data from which the energetics are determined. The inset shows the single exponential behavior of the excess damping. (c) Phase versus temperature for a single ^{3}He density of 0.046 Å^{-2} at four drive powers. Lines are drawn as a guide to the eye.

For low ^{3}He densities, p_1 takes the form of a Boltzmann distribution and for higher ^{3}He densities we use the Fermi distribution discussed earlier. By fitting the appropriate form of R_{ex} to the excess attenuation data, we can extract an activation energy, Δ' for each ^{3}He density. We show in the inset of Fig. 1(b) a plot of $\ln(R_{ex})$ vs $1/T$ for a ^{3}He density of 0.036 Å^{-2}. Although we might expect to see multiple exponential behavior since the ^{3}He is first excited into a higher-energy state in the film and then at higher temperatures is evaporated into the vapor, the data show only a single exponential. This indicates that the excess damping is more sensitive to the ^{3}He inside the ^{4}He film rather than the ^{3}He on top, as might be expected since the ^{3}He in the ^{4}He contributes to the normal component. The independence of the damping from evaporation into the vapor is also evident from the fact that at low temperatures the damping does not have any apparent dependence on ^{3}He density. For this reason we fit the single-exponential function for R_{ex} over the entire temperature range for all the ^{3}He densities. The excess damping data and typical fits are shown in Fig. 1(b).

An additional property of interest available from the third-sound measurements is the phase between the drive pulse and the ^{4}He film thickness above the driver. It is predicted by Bergman[7] that there should be a 180° phase difference between the temperature and the ^{4}He film thickness for third sound in pure ^{4}He films. This indicates that when the temperature of the driver is a maximum, the film thickness above it is a minimum. The quantitative relationship between temperature and film thickness oscillation predicted by Bergman was earlier measured[22] for third sound in a pure ^{4}He film. For the current measurement on mixture films, we see that within experimental error, for all the ^{3}He densities, at the lowest drive powers and below 200 mK there is indeed a 180° phase difference [one ^{3}He coverage is shown in Fig. 1(c)]. Then, abruptly near 200 mK, the phase changes to zero, indicating that the drive and response are in phase. Below 200 mK, the higher the drive power, the closer the drive and response are to being in phase. Above 200 mK there is no drive power dependence. This general behavior may be related to the curious pulse inversion seen in earlier third-sound work at much higher coverages.[22]

From the fits to the frequency data in Fig. 1 we derive the ground-state binding energy ε_0, and from the fits to the excess damping data we derive an activation energy Δ'. Along with the Fermi energies from NMR measurements, we can derive an excited-state energy ε_1 if we assume the activation energy Δ' to be related to Δ as $\Delta' = \Delta$. These two energies are compared in Fig. 2 to the ground-state and first-excited-state energies derived from NMR.[5] The agreement is good, which indicates that the assumption that we can ignore the higher-energy state in the film in the evaporation region is reasonable.

Also included in Fig. 2 is the theory of Anderson and Miller[23] for the ^{3}He density dependence of two energy states on a similar ^{4}He film ($d_4 = 3.4$ layers). They allowed for two states to be available in the film. We see reasonable agreement with their first excited state, especially at the higher densities, but poor agreement with the

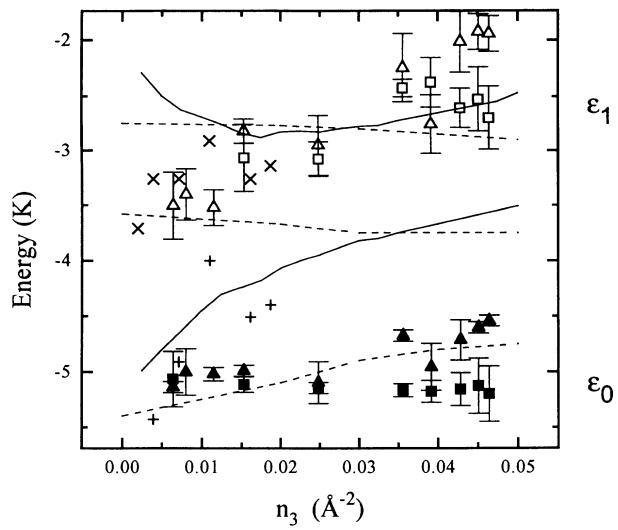

FIG. 2. Ground-state (solid symbols) and first-excited-state (open symbols) dependence on ^{3}He density of the binding energies of ^{3}He to a thin ^{4}He film. Energies from third sound (squares) are compared to the energies from NMR (triangles). Solid lines are theory of Anderson and Miller for a similar film. Dashed lines are theory of Pavloff and Treiner for ^{3}He on bulk ^{4}He. Crosses and pluses are data from Ref. 2 for a similar film.

ground state. Pavloff and Treiner[24] calculate the ^{3}He density dependence of the surface states on bulk ^{4}He using a density-functional approach; the lowest three of these are included in Fig. 2. Our ground-state energies are closer to those predicted by Pavloff and Treiner for ^{3}He on bulk ^{4}He than to those predicted by Anderson and Miller. For our ^{4}He coverage, which is slightly higher than Anderson and Miller's, we would expect to be closer to the binding energy of ^{3}He on bulk ^{4}He, but we also would expect that the binding energies *increase* towards this value as ^{4}He coverage is increased.[3] We are also in excellent agreement with a calculation of Pavloff and Treiner for a ^{3}He impurity on a ^{4}He film at our lowest ^{3}He coverage.[3] In this theory they take into account the proper ^{4}He density profile, and allow multiple states. They also note[1] that if, instead of bulk ^{4}He they have a film of ^{4}He their highest energy shown in Fig. 2 would be "pushed up" somewhat. It is possible that in our analysis we cannot distinguish between the higher two states, and are seeing an averaging of the two. Also included in Fig. 2 are the energies from the heat-capacity data of Bhattacharyya, DiPirro, and Gasparini[2] for similar film configuration ($d_4=3.4$ layers) on Nuclepore for some low ^{3}He densities; agreement is reasonable for both energy states.

In summary, we have observed the progression of the third-sound frequency, damping and phase as a function of temperature and ^{3}He density on a thin ^{4}He film. From the third sound, we have derived the absolute values of the ground- and first-excited bound state energies of the ^{3}He to the thin ^{4}He film as a function of increasing ^{3}He density, and observe that the agreement with measurements from NMR is good. The first-excited-state energy derived from third sound is in reasonable agreement with theory, although the ground state differs somewhat. There is an interesting feature observed in the phase of the third-sound mode—the relationship between the temperature fluctuations and crest height of the third-sound wave has an abrupt 180° phase change near the temperature where the ^{3}He starts to evaporate into a higher-energy state.

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