Third Sound in Layered Films: H↓-^{4}He and ^{3}He-^{4}He

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The (linearized) hydrodynamics of a layered film configuration is considered for propagating modes of third sound. The theory is applied to (normal) ^{3}He-^{4}He and H↓-^{4}He mixture film systems. When both films are superfluid, two propagating modes at each wave vector k will be observed. Thus, the detection of superfluid onset in the H↓ film should be direct and unambiguous. When the upper film is normal, an easily measurable shift in the ^{4}He third sound velocity, directly related to the physical characteristics of the normal film, will be observed.

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The suggestion of Stwalley and Nosanow[1] that our present day high-magnetic-field, low-temperature capabilities might be sufficient to form long-lived samples of (electron) spin-polarized hydrogen, H↓, has led to several recent experimental reports on this exotic system. Silvera and Walraven[2,3] and Cline *et al.*[4] reported success at maintaining samples with ~ 10^{17} atoms/cm^3 for times on the order of 10^4 s. Morrow *et al.*[5] have measured the binding energy of a hydrogen atom to a ^{4}He surface to be ≈ 0.9 K. The importance of this latter measurement lies in the crucial role that ^{4}He plays in the system. The walls of the sample cell are coated with a ^{4}He superfluid film whose purpose is to shield the H↓ atoms from the inevitable magnetic impurities in the walls. That is, one obtains a long-lived H↓ sample by minimizing the occurrence of "wrong" spin atoms whose presence leads to recombination (into the molecular, H$_2$, ground state) and degradation of the sample. As stressed by Silvera and Walraven,[2,3] the absence of the ^{4}He "shield" causes immediate degradation of the sample (indeed in their case, the H↓ mean free paths are macroscopic). Thus, Morrow *et al.*[5] were impelled to study in detail the interaction of a H atom with a ^{4}He surface. One may infer from the large binding energy which was obtained that the surface state can contain areal densities $\lesssim 10^{14}$ atoms/cm^2 which, as noted by Edwards and Mantz,[6] is sufficient to drive a superfluid transition (presumably of the Kosterlitz-Thouless-Nelson[7] type) at accessible temperatures.

In this Letter we shall present a theory which describes some of the hydrodynamic consequences of a layered (superfluid) film configuration. The modes of superfluid films are known as third sound[8] and we find that if the H↓ film goes superfluid on the already superfluid ^{4}He, see Fig. 1 (at temperatures of interest the ^{4}He film will always be below its onset), an easily observable pair of modes will appear. Moreover, the very presence of a H↓ "blanket" on the ^{4}He film causes a shift in the ^{4}He third sound speed irrespective of whether or not the H↓ is superfluid (this shift is directly related to the H↓ density). In the *normal* blanket limit our expression for the third sound shift has received experimental verification in the work of Ellis *et al.*[9] on ^{3}He-^{4}He mixture films.

We assume that a system with two fluid constituents forms as two films bound to a substrate and layered as shown in Fig. 1. The lower film will be taken to be superfluid ^{4}He and the upper film will be allowed to be either normal or superfluid H↓ with no mixing of the constituents.[10] The characterization of the H↓ one-atomic-layer-thick system as a film, although perhaps surprising, is perfectly reasonable in the light of the torsion oscillator results of Berthold, Bishop, and Reppy[11] for submonolayer ^{4}He films on Vycor. Thus, following the approach of Rutledge *et al.*[12] we characterize each fluid by its column height at each point along the substrate by introducing complex order parameters ψ_4 and ψ_1 defined by

$$|\psi_4|^2 = n_4 h_4(\vec{\rho}, t), \qquad |\psi_1|^2 = n_1 h_1(\vec{\rho}, t), \tag{1}$$

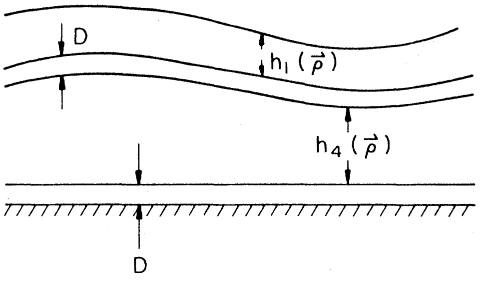

FIG. 1. Schematic drawing of a layered film system.

where n_4 (n_1) is the average number density in the ^{4}He (H$\downarrow$) film and $h_4(\vec{\rho},t)$ $[h_1(\vec{\rho},t)]$ is the height of the ^{4}He (H$\downarrow$) film at position $\vec{\rho}$ and time t. We may now construct the energy of the system in terms of the order parameters:

$$E[\psi_1,\psi_4]=E_4[\psi_4]+E_1[\psi_1,\psi_4]\,, \tag{2}$$

where

$$E_4[\psi_4]=\int d^2\rho\{(\hbar^2/2m_4)|\nabla\psi_4|^2-\mu_4|\psi_4|^2+n_4aW_4[\psi_4]\},$$

and

$$E_1[\psi_1,\psi_4]=\int d^2\rho\{(\hbar^2/2m_1)|\nabla\psi_1|^2-\mu_1|\psi_1|^2+n_1aW_1[\psi_1,\psi_4]\}.$$

In the above, μ_4 and μ_1 are the chemical potentials that determine the equilibrium structure of the films and W_4 (W_1) is the energy of the ^{4}He (H$\downarrow$) film in the external van der Waals potential. (We find it convenient to introduce the length scale a, defined by $n_4a^3=1$.) In the above we have neglected surface-tension contributions since they are unimportant in the long-wavelength limit. For the purposes of this discussion we treat the films as incompressible[13] by which we mean that the compressional velocities tend to be much larger than the velocities associated with the van der Waals potential (i.e., it is easier to change the column height at fixed density than it is to change the density at fixed column height). The van der Waals terms in Eq. (2) can be written

$$W_4[\psi_4]=-\alpha_4\int_D^{D+h_4(\vec{\rho},t)}\left(\frac{a}{z}\right)^3\frac{dz}{a}=\tfrac{1}{2}\alpha_4\left(\frac{a}{D}\right)^2\left[\left(\frac{D}{D+h_4}\right)^2-1\right], \tag{3a}$$

where α_4 is the He-substrate van der Waals parameter and the mobile portion of the ^{4}He film begins at $z=D$ (where $z=0$ is the plane of the substrate). Similarly, for the H$\downarrow$ film,

$$W_1[\psi_1,\psi_4]=-\alpha_1\int_{D+h_4}^{D+h_4+h_1(\vec{\rho},t)}\left(\frac{a}{z}\right)^3\frac{dz}{a}=\tfrac{1}{2}\alpha_1\left(\frac{a}{D}\right)^2\left[\left(\frac{D}{D+h_4+h_1}\right)^2-\left(\frac{D}{D+h_4}\right)^2\right]. \tag{3b}$$

The H$\downarrow$ fluid is driven by the substrate at both its upper and lower edges. The substrate pulls on a hydrogen column of height h_1 that rides on a helium column of height h_4. Thus, displacement of the ^{4}He surface is coupled to displacement of the H$\downarrow$ surface. For both films we introduce displacements, δh, from the equilibrium film heights, $\bar{h}$, defined by $h(\vec{\rho},t)=\bar{h}+\delta h(\vec{\rho},t)$. If we introduce the concomitant order-parameter fluctuations, $\delta\psi$, we have from Eq. (1)

$$n\bar{h}=\psi_0^2;\quad n\,\delta h=\psi_0(\delta\psi+\delta\psi^*)+|\delta\psi|^2. \tag{4}$$

We now substitute Eqs. (4) into the energy, Eqs. (2) and (3), and discard terms of higher than second order in $\delta\psi$. The vanishing of the linear term (the equilibrium condition) determines the film heights as functions of the chemical potentials. Thus,

$$\mu_4=-\alpha_4\left(\frac{a}{D+\bar{h}_4}\right)^3-\left(\frac{\alpha_1n_1}{n_4}\right)\left[\left(\frac{a}{D+\bar{h}_4+\bar{h}_1}\right)^3-\left(\frac{a}{D+\bar{h}_4}\right)^3\right];\quad \mu_1=-\alpha_1\left(\frac{a}{D+\bar{h}_4+\bar{h}_1}\right)^3. \tag{5}$$

The hydrodynamics of the coupled film system follows from the equations of motion

$$i\hbar\,\partial\psi_4/\partial t=\delta E/\delta\psi_4^*, \tag{6}$$

$$i\hbar\,\partial\psi_1/\partial t=\delta E/\delta\psi_1^*. \tag{7}$$

If we introduce dimensionless real and imaginary parts for the order-parameter fluctuation

$$\delta\psi\equiv(n\bar{h})^{1/2}(\eta+i\varphi), \tag{8}$$

then we find from Eq. (6)

$$\dot{\eta}_4=-(\hbar/2m_4)\nabla^2\varphi_4, \tag{9}$$

$$\hbar\dot{\varphi}_4=(\hbar^2/2m_4)\nabla^2\eta_4-2F_{44}\eta_4-2F_{41}\eta_1, \tag{10}$$

and from Eq. (7)

$$\dot{\eta}_1 = -(\hbar/2m_1)\nabla^2\varphi_1, \tag{11}$$

$$\hbar\dot{\varphi}_1 = (\hbar^2/2m_1)\nabla^2\eta_1 - 2F_{11}\eta_1 - 2F_{14}\eta_4. \tag{12}$$

In Eqs. (9)–(12), $\dot{x}$ denotes partial derivative of x with respect to time and the coupling constants are defined by

$$F_{44} = 3\alpha_4\left(\frac{\bar{h}_4}{a}\right)\left[\gamma\left(\frac{a}{D+\bar{h}_4+\bar{h}_1}\right)^4 + (1-\gamma)\left(\frac{a}{D+\bar{h}_4}\right)^4\right], \tag{13}$$

$$F_{11} = 3\left(\frac{\bar{h}_1}{a}\right)\alpha_1\left(\frac{a}{D+\bar{h}_4+\bar{h}_1}\right)^4, \tag{14}$$

where $n_4\bar{h}_4F_{41} = n_1\bar{h}_1F_{14} = n_1\bar{h}_4F_{11}$, and the dimensionless parameter $\gamma \equiv (\alpha_1 n_1/\alpha_4 n_4)$.

The coupled equations of motion, Eqs. (9)–(12), can be solved straightforwardly by Fourier transforms. We find

$$C_{3\pm}{}^2 = \tfrac{1}{2}\left\{[C_{34}{}^2(1+\Delta)+C_{31}{}^2] \pm [C_{34}{}^2(1+\Delta)-C_{31}{}^2]\left(1+\frac{4F_{41}F_{14}/m_4m_1}{[C_{34}{}^2(1+\Delta)-C_{31}{}^2]^2}\right)^{1/2}\right\}, \tag{15}$$

where the parameter Δ is defined by

$$\Delta = -\gamma[1-(D+\bar{h}_4)^4(D+\bar{h}_4+\bar{h}_1)^4], \tag{16}$$

$$C_{34}{}^2 \equiv 3\frac{\alpha_4}{m_4}\left(\frac{\bar{h}_4}{a}\right)\left(\frac{a}{D+\bar{h}_4}\right)^4$$

is the square of the pure-^{4}He third sound speed, and

$$C_{31}{}^2 \equiv 3\frac{\alpha_1}{m_1}\left(\frac{\bar{h}_1}{a}\right)\left(\frac{a}{D+\bar{h}_4+\bar{h}_1}\right)^4$$

is the square of the pure-H$\downarrow$ (in the sense that the lower film is normal) third sound speed. In Eq. (15), $C_{3\pm} = \omega/k$ is the third sound phase velocity and, in the long-wavelength limit, we have neglected the dispersive higher-order contributions in k. Thus, in the case when both films are superfluid there will be *two* modes at each value of k. The + mode is the shifted ^{4}He third sound velocity and the − mode is the shifted H$\downarrow$ third sound velocity. *Third sound detection should provide a clear and unequivocal demonstration of superfluidity in the H$\downarrow$ film.* The sensitivity of current third sound detectors makes it possible to see the H$\downarrow$(C_{3-}) mode down to relatively low H$\downarrow$-film areal densities. [For example, capacitive detection methods can easily sense a height change of 10^{-2} Å for a ^{4}He film with areal density ~ 10^{15} atoms/cm^2, so that for a H$\downarrow$ film with ~ 10^{13} atoms/cm^2 a change in height of 1 Å could be detected.] We note that the present results are obtained for $T = 0$ K; however, following Ref. 12, it is simple to account for finite temperature by calculating the momentum density due to the third sound quanta by using the standard[14] Landau quantum hydrodynamic recipe.

In the case where the upper film is normal, we set $C_{31} = 0$ in Eq. (15) to obtain

$$C_3{}^2 = C_{34}{}^2(1+\Delta), \tag{17}$$

where Δ is defined in Eq. (16). The ^{4}He third sound velocity is shifted by the factor Δ due to the normal-fluid blanket. This shift arises because the interface between the two films is driven by the energy difference that results from changes in the lower-film column height. If the upper film has the same interaction with the substrate as the lower film *and* the two films have the same number density, then changes in the location of the interface cost no energy and thus are not restored. An apparent example of a normal-fluid blanket is supplied by ^{3}He-^{4}He mixture films in the low-temperature limit. In work to be reported elsewhere, Ellis *et al.*[9] have found that Eq. (17), which describes the films in the completely phase-separated limit, yields excellent agreement with the experimentally measured third sound speeds—with no adjustable parameters. Thus, *by measuring the third sound shift, one is directly probing the characteristics of the (normal) H$\downarrow$ film.*

In summary, we have examined the hydrodynamic properties of a layered film configuration. For a normal-fluid blanket our results have received partial confirmation in studies of the ^{3}He-^{4}He mixture film system. If the upper film goes superfluid, we have assumed that its propagating modes are third-sound-like and have developed the consequences: Equation (15) shows two modes at each value of k. [Our assumption that the H$\downarrow$ normal fluid will "follow the motion" of the He normal fluid is based on extrapolating Osborne's[15] classic results for He vapor on the He liquid. If this should not be the case, the hydrodynamics will be somewhat more complicated.] In some very thin film geometries the H$\downarrow$ third sound speed may be nearly as large as the compressional speed. In this limit, one must take proper account of the compressibility of the H$\downarrow$ film. This is straightforward[16] and leads to a more complicated version of Eq. (15) whose basic physics is unchanged.

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[1] W. C. Stwalley and L. H. Nosanow, Phys. Rev. Lett. 36, 910 (1976).

[2] I. F. Silvera and J. T. M. Walraven, Phys. Rev. Lett. 44, 164 (1980).

[3] J. T. M. Walraven and I. F. Silvera, Phys. Rev. Lett. 44, 168 (1980).

[4] R. W. Cline, T. J. Greytak, D. Kleppner, and D. A. Smith, J. Phys. (Paris), Colloq. 41, C7-141 (1980).

[5] M. Morrow, F. Jochemsen, A. J. Berlinsky, and W. N. Hardy, Phys. Rev. Lett. 46, 195 (1981).

[6] D. O. Edwards and I. B. Mantz, J. Phys. (Paris), Colloq. 41, C7-257 (1980).

[7] D. R. Nelson and J. M. Kosterlitz, Phys. Rev. Lett. 39, 1727 (1977).

[8] K. R. Atkins, Phys. Rev. 113, 962 (1959).

[9] F. M. Ellis, R. B. Hallock, M. D. Miller, and R. A. Guyer, Phys. Rev. Lett. 46, 1491 (1981).

[10] The zero-concentration chemical potential for a H atom in ^{4}He has been calculated to be ~40 K, M. D. Miller, Phys. Rev. B 18, 4730 (1978).

[11] J. E. Berthold, D. J. Bishop, and J. D. Reppy, Phys. Rev. Lett. 39, 348 (1977).

[12] J. E. Rutledge, W. L. McMillan, J. M. Mochel, and T. E. Washburn, Phys. Rev. B 18, 2155 (1978).

[13] For a discussion of the effects of compressibility on third sound, see R. D. Puff and J. G. Dash, Phys. Rev. B 21, 2815 (1980). (See also the discussion in the last paragraph of this paper.)

[14] See, e.g., J. Wilks, *Liquid Helium* (Oxford Univ. Press, New York, 1967).

[15] D. V. Osborne, Proc. Phys. Soc., London 80, 103, 1343 (1962).

[16] R. A. Guyer and M. D. Miller, unpublished.

Calorimetric Investigation of Spin-Glass Ordering in CuMn

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New measurements near T_{sg} characterize the heat-capacity anomaly associated with spin-glass ordering. There are no discontinuities in the heat capacity or its temperature or field derivatives but there is a relatively sharp anomaly in the field derivative and a broad anomaly in the temperature derivative. A failure of thermodynamics to predict correctly the relation between heat capacity and magnetic susceptibility may be a manifestation of the "nonergodic" behavior of disordered systems.

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The nature of the transition to the ordered state in metallic spin-glasses continues to be an interesting and challenging aspect of the more general problem of disordered systems. The discovery[1] of a sharp cusp in the low-field ac susceptibility, χ_{ac}, at a characteristic temperature, T_{sg}, led to a renewal of both experimental and theoretical investigations of spin-glasses, and, together with Mössbauer[2] and magnetic remanence[3] data, prompted consideration of the possibility of a thermodynamic phase transition at T_{sg}. Edwards and Anderson (EA) proposed a model in which the long-range Ruderman-Kittel-Kasuya-Yosida interaction between randomly distributed spins was replaced by a Gaussian distribution of bond strengths coupling spins on a regular lattice.[4] Mean-field solutions of this model and extensions of it do produce a cusp in χ but they also predict a cusp in the magnetic specific heat, C. Although a broad anomaly that extends from the lowest temperatures at which C has been measured to well above T_{sg} is well known, no feature of the type predicted had been reported. More recent theoretical work has addressed both the essential