1981

Phase Separation in 3He-4He Mixture Films

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models for $J_{nn}=0$. For $J_{nnn}>0$, the system orders into one of $q(q-1)$ degenerate ground states with both sublattices ordered ferromagnetically but antiferromagnetically with respect to each other. Figure 4 shows the average energy versus the temperature for $J_{nnn}=-J_{nn}$ and three values of q. In all cases, the system disorders via two first-order transitions. Pronounced hystersis effects are observed especially for higher values of q. Between the two transition temperatures the system is found to be in a BSS state characterized by one of the sublattices being ordered ferromagnetically in one of the q states and the other sublattice populated randomly with the remaining $q-1$ states. This leads to a large gain in entropy with almost all the nn bonds satisfied and some of the nnn bonds being unsatisfied. The results for $q \geq 5$ differ from those discussed above for $q=4$ only in the magnitude of the associated latent heat. This BSS state has also been observed in three dimensions.

It is a pleasure to thank L. Kadanoff, M. den Nijs, D. Fisher, P. Hohenberg, J. Hirsch, S. Shenker, and C. Varma for many stimulating discussions and J. Cardy for communicating his results prior to publication. This work was supported in part by the National Science Foundation under Grants No. DMR-80-17758, No. DMR-79-07232, and No. DMR-78-24698.

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[1]B. Nienhuis, A. N. Berker, E. K. Riedel, and M. Schick, Phys. Rev. Lett. 43, 737 (1979); M. P. M. den Nijs, J. Phys. A 12, 1857 (1979); M. Nauenberg and D. J. Scalapino, Phys. Rev. Lett. 44, 837 (1980); C. Rebbi and R. H. Swendsen, Phys. Rev. B 21, 2094 (1980); L. P. Kadanoff, Phys. Rev. B 22, 1405 (1980).

[2]A. N. Berker and L. P. Kadanoff, J. Phys. A 13, L259 (1980).

[3]J. R. Banavar, G. S. Grest, and D. Jasnow, Phys. Rev. Lett. 45, 1242 (1980).

[4]R. J. Baxter, J. Math. Phys. (N.Y.) 11, 3166 (1970).

[5]J. V. José, L. P. Kadanoff, S. Kirkpatrick, and D. R. Nelson, Phys. Rev. B 16, 1217 (1977).

[6]J. L. Cardy, to be published.

[7]J. Hirsch, private communication.

[8]M. P. M. den Nijs, private communication.

[9]R. J. Baxter, J. Phys. C 6, L445 (1973).

[10]L. P. Kadanoff and A. Brown, Ann. Phys. (N.Y.) 121, 318 (1979).

Phase Separation in Films of ^{3}He-^{4}He Mixtures

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(Received 9 January 1981)

We report detailed observations of the propagation of third sound in ^{3}He-^{4}He mixture films as a function of temperature and ^{3}He concentration. The data are consistent with a simple model for the film and we conclude that thin ^{3}He-^{4}He mixture films exhibit nearly complete phase separation.

PACS numbers: 67.60.-g, 67.40.Pm

The study of thin ^{4}He films[1,2] is at the forefront of the investigation into the physics of (quasi-) two-dimensional systems. In particular, the agreement achieved between experiment[1] and the Kosterlitz-Thouless-Nelson theory[3] of superfluid onset is a major triumph for that vortex-unbinding picture. It is expected that the addition of a ^{3}He component will, in analogy to the bulk, make this rich system even richer. There has already been some theoretical[4] and experimental[5,6] work concerned with the effects of the ^{3}He impurity on superfluid onset. The basic thrust of this paper, however, pertains to the phase-separation aspects of the equation of state. Below we shall present third-sound measurements in the mixture together with a hydrodynamic analysis which will lead us to conclude that the state of the film (for $T \lesssim 0.5$ K) is one of layered phase separation.

In the general situation we can picture the films as shown in Fig. 1. The lower film (l) will contain a (film-averaged) mass concentration, x_{3l} of ^{3}He, and in addition will contain *all* the superfluid. The upper film (u) will be considered to be

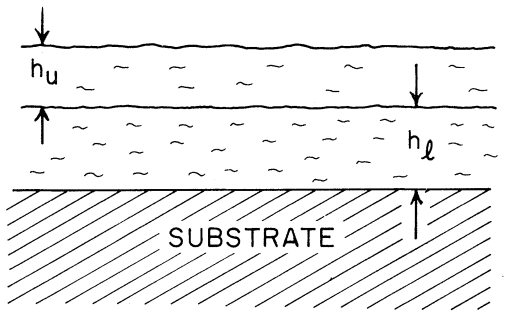

FIG. 1. Schematic representation of the general two-layer situation for the mixture film.

a normal fluid blanket of ^{3}He and ^{4}He. (A limiting case naturally has all the atoms in the lower film completely mixed but, due to the external field, not homogeneous.) The linearized hydrodynamic equations are[7] (after neglecting dissipative terms)

$$\partial\rho_l/\partial t + \nabla\cdot\vec{g} = 0, \tag{1}$$

$$\partial\rho_{3l}/\partial t = 0, \tag{2}$$

$$\partial\sigma_l/\partial t = 0, \tag{3}$$

$$\partial\vec{v}_s/\partial t + \nabla(\mu - \Delta x_{3l}) = 0, \tag{4}$$

where ρ_l is the mass density, $\vec{g} = \rho_s\vec{v}_s$ is the momentum density, σ_l is the entropy density, μ is the chemical potential per unit mass, $\Delta = \mu_3 - \mu_4$, and we have set $\vec{v}_n = 0$ everywhere. In the following we shall treat the films as incompressible and we shall neglect the entropy conservation equation, Eq. (3) (this is permitted since we find empirically that the data in the range of concern contain only a negligible temperature dependence). Then, following standard treatments,[8] Eqs. (1), (2), and (4) yield for the third-sound velocity in the long-wavelength limit

$$C_3^2 = \frac{\langle\rho_s\rangle}{\rho_l} h_l \left(1 + \frac{x_{3l}}{\rho_l}\frac{d\rho_l}{dx_{3l}}\right)\left\{-f_l(h_l) + \frac{\rho_u}{\rho_l}[f_u(h_l) - f_u(h_l + h_u)]\right\}, \tag{5}$$

where $\langle\rho_s\rangle$ is the film-averaged superfluid density and the f's are the effective forces on the lower or upper film at the interface and upper surface. We note that, since there is no superfluid in the upper film, it is constrained to move rigidly with the lower surface. (If the upper film were fixed in space, then it would act as a second substrate—a situation for which there is no obvious evidence.) The experiments which we shall describe below were performed with a fixed coverage of ^{4}He and varying amounts of ^{3}He. It is thus convenient to divide Eq. (5) by $C_{30}^2 \equiv (\langle\rho_{s4}\rangle/\rho_4)h_4 f_4(h_4)$ the third-sound velocity squared of the pure ^{4}He component. Equation (5) can then be written

$$\frac{C_3^2}{C_{30}^2} = \left(\frac{m_4}{m_l}\right)\left(\frac{h_4}{h_l}\right)^4\left(1 - \frac{n_u}{n_l} + \frac{n_u/n_l}{(1 + h_u/h_l)^4}\right), \tag{6}$$

where m_l is the mass per particle in the lower film, and n_u and n_l are the average *number* densities in the upper and lower films, respectively. In achieving Eq. (6) we have neglected *excess* volumes in the lower film and have also assumed that the number of superfluid atoms at a given temperature remains constant as one adds ^{3}He. We also use the standard form $f(h) = -(3\alpha/m)h^{-4}$, where α is the van der Waals parameter.

The experiments were conducted in a beryllium-copper chamber equipped with a superfluid valve and coupled by a weak thermal link to a recirculating ^{3}He refrigerator. (See Fig. 2.) The experimental chamber was equipped with a pressure gauge for *in situ* vapor-pressure measurements, a parallel-plate capacitor for direct adsorbed helium measurements and superconducting aluminum strips on glass which served to detect both third sound in the helium film and ordinary sound in the vapor above the film. The ordinary sound velocity measurements allowed a direct determination[9] of the ^{3}He concentration in the vapor mixture in the chamber. The chamber was equipped with 798 glass plates 18 mm in diameter and 25 μm thick, rather than Al_2O_3, to provide adequate surface area to stabilize the film thickness and suppress the effects of capillary condensation. Capillary condensation was demonstrably absent[6] from the apparatus for the results we report here.

In these studies a known amount of ^{4}He was admitted to the chamber at 0.4 K. The ^{4}He so admitted was adequate to provide a film thickness on the glass of $h_4 = 5.7$ atomic layers at low temperatures. The superfluid valve was sealed and the third-sound velocity, pressure and adsorbed helium were subsequently measured as a function of increasing temperature. Sequentially, varying amounts of ^{3}He were then added at 0.4 K and after each increase in the amount of ^{3}He in the cell the measurements were repeated as a function of increasing temperature. We report our observations of the third-sound velocity as a

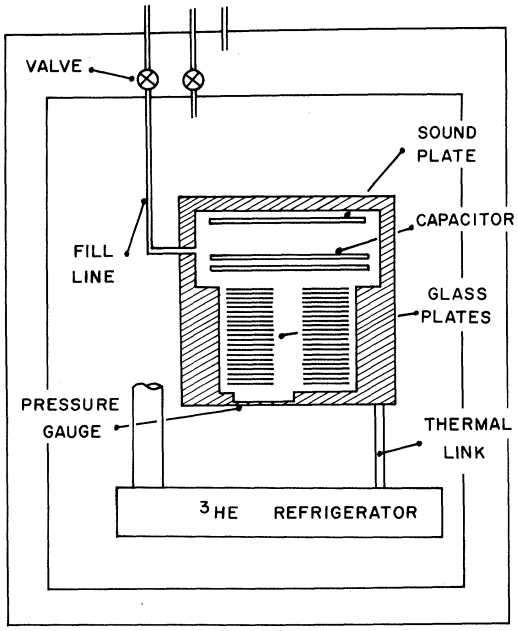

FIG. 2. Schematic representation of the apparatus used for the present measurements.

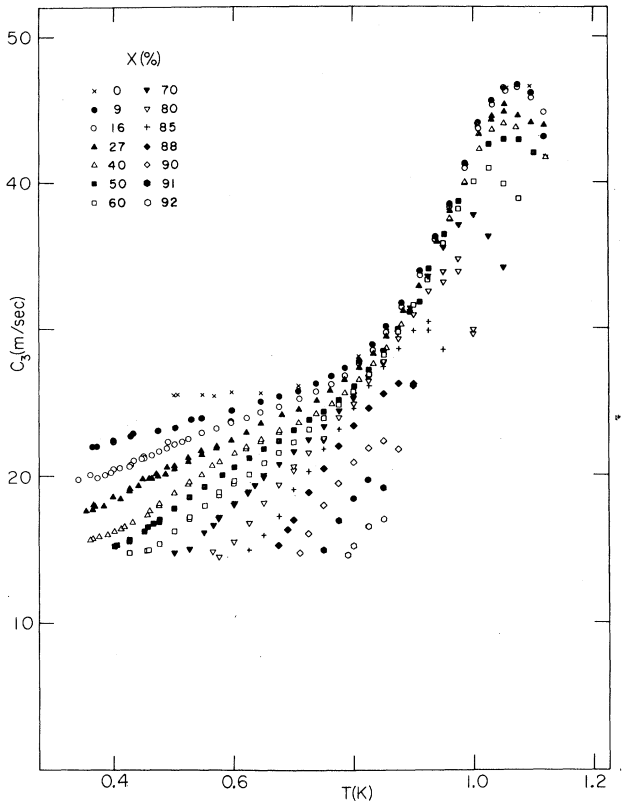

FIG. 3. The observed velocity of third sound in the experimental apparatus as a function of temperature for various values of the number concentration X. We define $X = N_3/(N_3 + N_4)$, where N_3 and N_4 are the numbers of ^{3}He and ^{4}He atoms admitted to the chamber, respectively.

function of number concentration X and temperature in Fig. 3. In all cases the addition of ^{3}He results in a reduction in the third-sound velocity and a depression of superfluid onset to lower temperatures. Since the experiments for each X were conducted in a sealed cell as a function of temperature, the film thickness, average film ^{3}He concentration, etc., do not remain constant along the curves of fixed X presented in Fig. 3.

For temperatures below about 0.7 K the amount of ^{4}He in the film is independent of temperature and remains fixed at the equivalent of 5.7 pure layers. The presence of a fixed amount of ^{4}He in the film makes the low-temperature data particularly interesting. In Fig. 4, we show C_3^2/C_{30}^2 as a function of the ratio of amount of ^{3}He, h_3, to the amount of ^{4}He (measured in layers $\equiv 3.6$ Å with $h_4 = 5.7$ layers). There are three sets of experimental points shown which correspond to data taken at $T = 0.40$, 0.45, and 0.50 K. It is one purpose of this paper to point out that in the limit of complete phase separation (i.e., $h_u = h_3$) Eq. (6) fits all the data to within 5% with *no* adjustable parameters. This solution is shown as the solid line in Fig. 4. Given the detail available to us concerning the conditions in the experimental cell, we have confidence in the comparison. The opposite limit of total mixing is also shown in Fig. 4 and it is evident that such a model can*not* fit the data.

The intermediate case of partial mixing (which *must* to some degree be the state at finite temperature) is rather complicated; however, the following points can be made: (1) The partially mixed state does not necessarily lie *between* the two extremes in Fig. 4 because of the strong dependence of C_3 on film thickness. (2) The data are not compatible with a model which fixes the number concentration in either the upper or lower film. (For example, if one wished to fix the upper-film concentration at 90%, then one would exhaust the ^{3}He below $\sim 0.65\, h_3/h_4$.)

The residuals generated by this model (i.e., the difference between the completely phase-separated calculation and the experimental data for the third sound) are rather small but nonetheless interesting because the calculated third-sound speeds are generally *slower* than the experiment requires. There are two very simple ways that

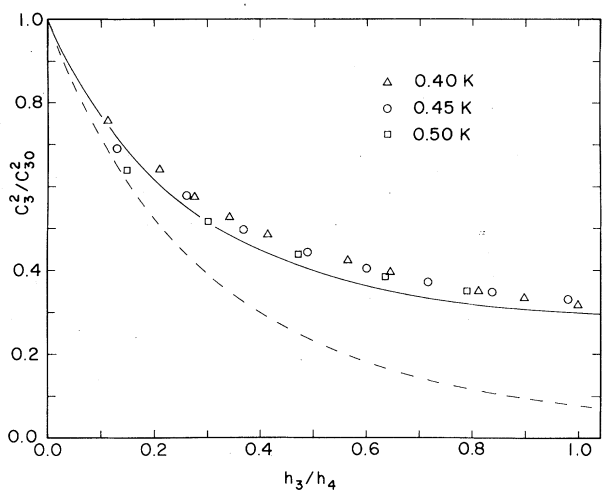

FIG. 4. The ratio of the square of the third-sound velocity to the square of the third-sound velocity for the case of pure ^{4}He as a function of the thickness of the added ^{3}He at several temperatures. The curves represent the predictions for complete phase separation (solid line) and perfect mixing (dashed line).

these computed third-sound speeds can be increased: Either increase the "restoring" force or make the film thinner. The former case can be realized if the ^{3}He blanket acted as a second substrate. One would then expect the residual to increase to an asymptotic value as the ^{3}He blanket became thick. The residuals, however, generally decrease as h_3/h_4 increases. The second somewhat speculative possibility is that we are witnessing the ^{3}He change its environment from being trapped in a surface state for $h_3 \lesssim 1$ layer to forming a well-defined film and concomitant interface for $h_3 > 1$ layer. That is, if the superfluid order parameter should respond to a filled ^{3}He surface state by receding by half a layer and then relaxing back to its original (pure ^{4}He) level as the ^{3}He is promoted out of the surface state into its own upper film then the qualitative behavior of the data can be understood.

In summary, we have reported low-temperature third-sound measurements in mixture thin films. We have extended the standard ^{4}He superfluid hydrodynamics to the case of low-temperature, long-wavelength third sound in the mixture. The agreement between the prediction [e.g., (6)] and the experimentally determined parameters is excellent. We thus find that the state of the film is one of phase separation. This conclusion is in disagreement with that of LaHeurte, Noiray, and Romagnan,[10] who conclude that phase separation is suppressed in thin mixture films. The phase separation we observe is not of the lateral or two-dimension type treated by the various works of Ref. 5; rather, we observe a layered film configuration. This layered structure is apparently driven by the external van der Waals field; perhaps analogous to the bulk phase separation in the presence of gravity. Thus, for films thicker than one mobile layer it may be true that layered phase separation is the natural state and lateral phase separation quite unlikely. Some expansion of the observations of Bishop and Reppy[11] might be relevant to the lateral case.

We have benefited from preliminary experimental work in collaboration with J. S. Brooks. We acknowledge a grant of computer time from the University Computer Center. This work was supported by the National Science Foundation.

[1]I. Rudnick, Phys. Rev. Lett. 40, 1454 (1978); D. J. Bishop and J. D. Reppy, Phys. Rev. Lett. 40, 1727 (1978).

[2]M. Chester and L. C. Yang, Phys. Rev. Lett. 31, 1377 (1973); J. G. Dash, Phys. Rev. Lett. 41, 1178 (1978); D. T. Ekholm and R. B. Hallock, Phys. Rev. Lett. 42, 449 (1979); B. Ratnam and J. Mochel, J. Low Temp. Phys. 3, 239 (1970).

[3]J. M. Kosterlitz and D. J. Thouless, J. Phys. C 6, 1181 (1973); D. R. Nelson and J. M Kosterlitz, Phys. Rev. Lett. 39, 1201 (1977).

[4]J. L. Cardy and D. J. Scalapino, Phys. Rev. B 19, 1428 (1979); A. N. Berker and D. R. Nelson, Phys. Rev. B 19, 2488 (1979); K. K. Mon and W. F. Saam, to be published.

[5]E. Webster, G. Webster, and M. Chester, Phys. Rev. Lett. 42, 243 (1979).

[6]F. M. Ellis, J. S. Brooks, and R. B. Hallock, in *Ordering in Two Dimensions*, edited by S. K. Sinha (Plenum, New York, 1980), p. 275.

[7]I. M. Khalatnikov, *Introduction to the Theory of Superfluidity* (Benjamin, New York, 1965).

[8]D. Bergman, Phys. Rev. 188, 370 (1969), and Phys. Rev. A 3, 2058 (1971); R. D. Puff and J. G. Dash, Phys. Rev. B 21, 2815 (1980).

[9]D. T. Ekholm and R. B. Hallock, Phys. Rev. B 21, 3913 (1980).

[10]J. P. LaHeurte, J. C. Noiray, J. P. Romagnan, and H. Dandache, Phys. Rev. B 22, 4307 (1980).

[11]D. J. Bishop and J. D. Reppy, Phys. Rev. B 22, 5171 (1980), Sec. VI.