Anomalous Wetting of Helium on Cesium

K. S. Ketola, S. Wang, and R. B. Hallock
Laboratory for Low Temperature Physics, Department of Physics and Astronomy, University of Massachusetts, Amherst, Massachusetts 01003

(Received 21 October 1991)

We report studies of the anomalous wetting of a cesium substrate by a liquid-helium film by means of the technique of third sound. In an apparatus which incorporates both glass and cesium substrates, we observe third sound to propagate across cesium for helium films such that the film thickness on glass is $d_g \gtrsim 11.5$ layers. For $d_g \lesssim 9$ layers, third sound does not propagate across the cesium substrate. The transition region is hysteretic as a function of the amount of helium in the experimental cell.

PACS numbers: 67.70.+n

Recently Cheng *et al.* [1] undertook a theoretical study of the behavior of helium in the vicinity of alkali-metal surfaces at $T=0$ K. Such surfaces are interesting since the potential well which binds the helium to the surface is extremely weak. This is particularly so for the case of the helium-cesium interaction; it is predicted [1] that helium will not wet cesium for any value of the chemical potential (at $T=0$ K).

In an attempt to study the adsorption of helium to alkali metals and to experimentally test the prediction of Cheng *et al.* [1] for cesium, we have used waves on a helium film, known as third sound [2], as a probe. Third sound is a long-wavelength disturbance which propagates on a liquid-helium film at temperatures below the superfluid transition. As is the case for a tidal wave on the ocean, the wave velocity is given by $v^2 \sim fH$, where f is the restoring force per unit mass and H is the depth of the medium. For third sound, f is due to the van der Waals force between the helium and the substrate. A more complete expression for the velocity of third sound, C_i, on surface i can be written as a function of the film thickness d_i by use of the relation

$$C_i^2 \simeq \langle \rho_s \rangle k_B \alpha_i \beta_i (3\beta_i + 4d_i) \frac{(1+TS/L)^2}{\rho m_4 d_i^3 (d_i+\beta_i)^2},$$

where $\langle \rho_s \rangle / \rho$ is the effective superfluid fraction [3] in the film, k_B is the Boltzmann constant, m_4 is the mass of a ^{4}He atom, L is the latent heat, S is the entropy, and α_i and β_i are parameters of the helium-substrate interaction. The passage of a third-sound wave crest (trough) is accompanied by a transient decrease (increase) in the local temperature of the film. This transient change in the ambient conditions can be observed by either capacitance [4] or thermometric [5] techniques.

Our experiments are conducted on a substrate, shown schematically in Fig. 1, made of borosilicate glass onto which four detectors and a generator of third sound have been deposited by vacuum evaporation. A resistive silver third-sound generator Z is located approximately at the center of the glass substrate. Four aluminum "detectors" of third sound (A,B,C,D) are located at symmetric positions on the substrate. These detectors operate as sensitive superconducting transition edge bolometers and measure the temperature swing associated with the passage of the third-sound wave. A region between detectors A and B has been modified by the application of a cesium film to the glass surface by *in situ* vacuum evaporation. The evaporation source is a commercial alkali-metal dispenser [6] which was located ~3.5 cm from the substrate. A collimator enclosed the dispenser and extends to 1 mm from the glass substrate so as to prevent the deposit [7] of cesium on the glass everywhere except for a well-defined region between detectors A and B. As deduced by third-sound measurements, cesium does not reach the regions B–D. We are unable to directly determine the thickness l of this cesium film, but from the parameters of the evaporation process we estimate l to be approximately 3 μm. The substrate is enclosed in a sealed brass chamber which is attached to the end of a standard cryostat insert and housed in a pumped helium bath. The amount of helium in the chamber can be changed by use of a filling capillary which enters the cell. The pressure P in the cell is kept below the saturated vapor pressure P_0 at the temperature T so that no bulk puddle of liquid exists; thus, the film is never in coexistence with bulk liquid. Under such conditions, the film thickness d_i on a given substrate (which the helium wets) can be related to P_0/P by the approximate relation $d_i^3(1+d_i/\beta_i) \simeq \alpha_i/T \ln(P_0/P)$. For glass $\alpha_g \simeq 27$ layers^{3}K and $\beta_g \simeq 42$ layers. For cesium these parameters have not been measured, but it is predicted [8] that $\alpha_{\rm Cs} \approx 0.5\alpha_g$. The film thickness values we

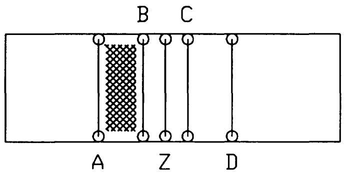

FIG. 1. Schematic of the glass substrate geometry used for this work. Z is a generator of third sound. The shaded region is an evaporated cesium film. The region between C and D has no evaporated coating; it is clean glass. Except for the cesium coating the substrate is symmetric about the driver Z.

report here are the values of the thickness on the clean glass substrate d_g as deduced by measurements of the third-sound velocity on the clean glass.

Third sound is generated thermally by the application of a current pulse to the driver Z. We use a square-wave drive with pulse widths in the vicinity of 50 μsec and are able to vary the amplitude A_3 of the third-sound pulse by varying the voltage amplitude V of the square-wave drive. For all of the measurements we report here, the third sound is linearly dependent [9] on the applied power. All four detectors of third sound have approximately the same sensitivity, and resistance calibrations are carried out over a temperature range of several tens of mK near the operating temperature of 1.38 K.

Data for two values of the helium film thickness taken at $T=1.38$ K as a function of increasing amounts of helium in the experimental cell are shown in Figs. 2 and 3. Figure 2(a) shows the change in the temperature of detector A as a function of time due to a third-sound pulse created at Z at $t=0.075$ msec for $d_g=11.5$ layers. The arrival of third sound at detector A at $t=3.3$ msec demonstrates that the cesium surface is covered by a helium film for this case. Figure 2(b) shows the direct third-sound pulse at $t=1.9$ msec for the helium-on-glass trajectory Z-D followed by an inverted pulse which has been reflected from the edge of the cesium nearest to Z and received at detector D. We are certain of the identity of the pulses due to the presence of detectors B and C which we also monitor. Note that the reflected signal has an amplitude which is negative compared to that of the direct signal Z-D. We have studied helium film thickness values as large as $d_g \simeq 30$ layers and, consistent with the behavior seen in Fig. 2, for $d_g \geq 11.5$ layers third sound propagates across the cesium.

Thinner films show an *entirely different* behavior. Figure 3 shows data taken at 10.7 layers. The striking observation here [Fig. 3(a)] is the absence of any signal at detector A. Also, notice in Fig. 3(b) that the amplitude of third sound reflected from the edge of the cesium and received at detector D has reversed sign. Data taken as a function of increasing film thickness at film coverages in the range $4.0 \leq d_g \leq 10.7$ are consistent with those shown in Fig. 3(a); third sound does not appear to propagate on cesium for films such that $d_g \leq 10.7$ layers.

We interpret the absence of third sound for small values of the chemical potential ($d_g \leq 10.7$ layers) to indicate that the helium film does not wet [10] the cesium with an increase in chemical potential in the same

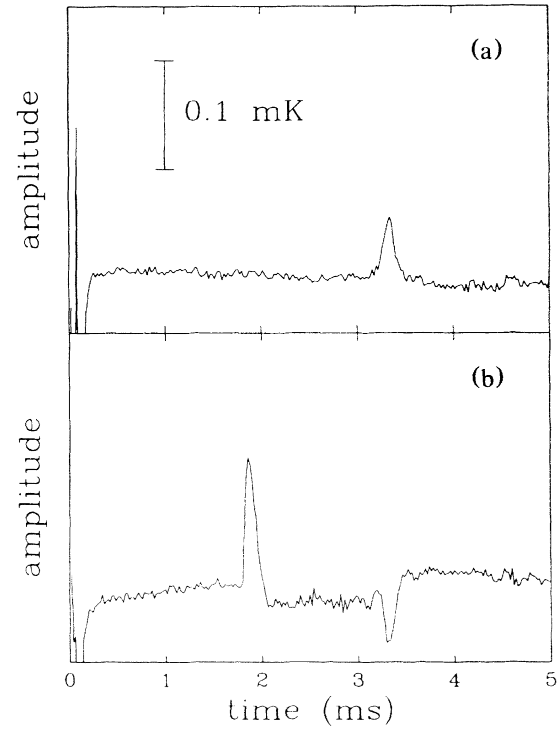

FIG. 2. Examples of third-sound pulses received at detectors A and D where the chemical potential is such that the thickness of a helium film on glass is $d_g=11.5$ atomic layers. (a) Third sound which has arrived at detector A after passing over the cesium surface. (b) Third sound observed at detector D. Note the presence of the direct pulse followed by the arrival of a pulse which has reflected from the edge of the cesium region. This reflected pulse arrives with an amplitude which is negative relative to that of the direct pulse.

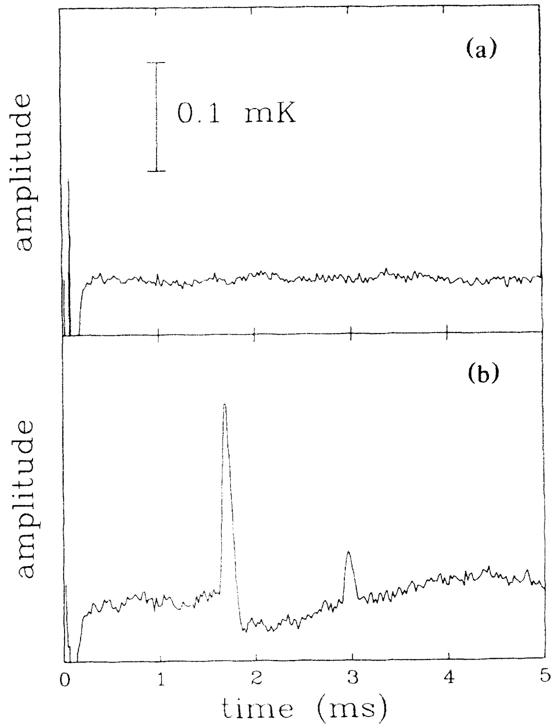

FIG. 3. Similar to Fig. 2 except $d_g=10.7$ layers. Note in (a) the absence of any direct third-sound pulse; the third sound does not propagate across the cesium for this case. Note also that the pulse reflected from the edge of the cesium region visible in (b) is of opposite sign to the case of Fig. 2(b).

manner as it does on nonalkali substrates. We believe that the possibility that helium does wet the cesium but that the superfluid density is zero in this range of chemical potential can be excluded for the following reason. A film which wets cesium is predicted [8] to have a thickness $d_{\mathrm{Cs}} \approx 0.78 d_g$ with an "immobile" helium layer adjacent to the substrate that is substantially thinner than is the case for glass. Thus, if the helium wets the cesium, the film thickness should be much greater than that necessary to satisfy the Kosterlitz-Thouless [11] criterion for superfluidity at 1.38 K and superfluid should be present with an appreciable superfluid fraction. The abrupt change in behavior for $d_g \approx 11$ layers indicates that there may be a prewetting (or percolation) transition at the value of the chemical potential represented by $d_g \approx 11$ layers.

For increasing values of the chemical potential, the appearance of third sound at detector A is an abrupt event. This can be seen in Fig. 4(a) which shows the amplitude of a third-sound pulse recorded at detector A, A_a, normalized by the amplitude recorded at detector D, A_D, vs d_g for increasing (solid symbols) and decreasing values of d_g. For decreasing chemical potential there is no abrupt extinction of the third-sound signal; A_a/A_D falls gradually with decreasing d_g. The evolution of the amplitude of third sound received at detector A is clearly hysteretic. The normalized amplitude of the third sound reflected from the edge of the cesium region closest to Z and received at detector D, A_r/A_D, bears an interesting relationship to the amplitude observed at detector A; this is shown in Fig. 4(b). For increasing values of d_g (solid symbols), A_r/A_D passes through zero and changes sign at the value of d_g at which third sound first appears at detector A. For decreasing values of d_g, a third-sound signal is observed at detector A for coverages d_g well below that at which A_r/A_D has passed through zero and reversed sign. Behavior similar to that shown in Figs. 2-4 has also been seen at $T = 1.48$ K.

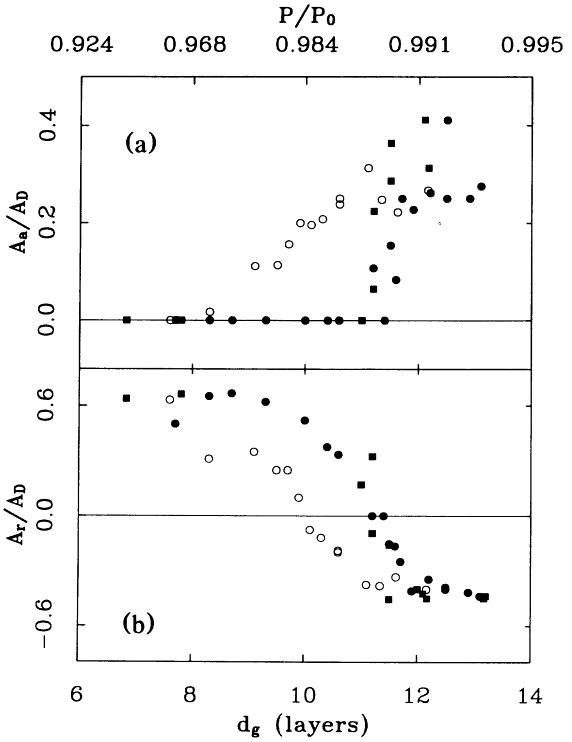

FIG. 4. Behavior of the third-sound pulse amplitudes as a function of d_g. Data (normalized to the amplitude of the direct third sound received at detector D, A_D, for the trajectory Z-D) are shown for increasing (solid symbols) and decreasing (open circles) values of d_g. (a) Amplitude ratio A_a/A_D, where A_a is the amplitude observed on detector A. (b) Amplitude ratio A_r/A_D, where A_r is the amplitude of third sound reflected from the edge of the cesium region closest to Z as recorded at detector D.

As evident from Fig. 2, the time of flight on the third sound on the cesium-coated substrate is larger than the time of flight on glass. This reduced velocity can be characterized by an index of refraction $n = C_g/C_{\mathrm{Cs}}$. As a result of the choice of the dimensions of the cesium region of our substrate and the relative locations of the third-sound generators and detectors, the index of refraction can be expressed as $n = 2\tau_a/\tau_D - 1$, where τ_a and τ_D are the measured times of flight for the trajectories Z-A and Z-D, respectively. We find n to be a decreasing function of d_g, falling from $n \sim 3$ for $d_g \sim 9$ to $n \sim 2.3$ for $d_g \sim 14$. This observed index of refraction could be due to a reduced value of the superfluid fraction on the cesium; but this is unlikely if the film wets. The reduced velocity on the cesium could also be due to the presence of a film on the cesium which is thicker than that on glass at the same chemical potential. However, this is unlikely for a flat surface since the van der Waals constant on glass is considerably larger than is expected [6] to be the case on cesium. Most likely, the enhanced time of flight is due to roughness [12] of the evaporated cesium substrate. Wetting behavior is predicted to be changed by the presence of roughness on a surface [13]; for the thinnest films the liquid-substrate interaction dominates the behavior, while for thicker films, the tendency to wet is enhanced by the topology of the surface structure. In this case, the effect of surface tension becomes more important.

Nacher and Dupont-Roc [14] very recently have studied the effective thermal conductance of a helium film contained within a sealed glass tube which has a ring of alkali metal on the glass at one spot along its length. They interpret their experimental results as evidence that the helium wets all of the alkali-metal surfaces studied *except* for cesium. In the experiments of Nacher and Dupont-Roc the film in the glass tube was always in coexistence with bulk liquid (and thus was saturated) whereas our films are never in coexistence with bulk liquid. It is also likely that the smoothness and impurity content of the cesium may be different in the two experiments.

At the conclusion of these measurements, an amount of oxygen was admitted to the sample cell adequate to completely oxidize the cesium and the apparatus was brought to room temperature. The cesium surface remained exposed to oxygen at 25 Torr at room temperature for $\sim 1\frac{1}{2}$ days. Subsequent measurements at 1.38 K for d_g in the range $3.3 \leq d_g \leq 14.6$ layers resulted in the observation of third sound at detector A for all coverages studied. The oxygen appears to have reacted with the cesium allowing the resulting surface to be wet by helium for all values of the chemical potential studied. Although the character of the surface following the oxygen dose is not well known, two observations have been made. The index of refraction for the oxygen-exposed surface was $n \sim 1$ over the coverage range studied and the third-sound pulses for this surface appear as those on glass for $4 \leq d_g \leq 8.5$, but appear wider than those on glass for $d_g \geq 8.5$.

We conclude that, for small values of d_g, helium does not wet cesium as evidenced by the absence of third-sound propagation across an evaporated cesium film. For thicker films, third sound is observed to propagate across this cesium surface. The transition region between the presence and absence of third sound on cesium is observed to be hysteretic. Further experimental work will be required to establish whether this apparent prewetting behavior [1,15] is induced by surface roughness.

This work was supported by the National Science Foundation through Grant No. DMR88-20517.

---

[1] E. Cheng, M. W. Cole, W. F. Saam, and J. Treiner, Phys. Rev. Lett. **67**, 1007 (1991); (private communication).

[2] K. Atkins and I. Rudnick, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland, Amsterdam, 1970), Vol. 6, p. 1.

[3] See, for example, S. Putterman, *Superfluid Hydrodynamics* (North-Holland, Amsterdam, 1974).

[4] J. S. Brooks, F. M. Ellis, and R. B. Hallock, Phys. Rev. Lett. **40**, 240 (1978); F. M. Ellis, J. S. Brooks, and R. B. Hallock, Rev. Sci. Instrum. **52**, 1051 (1981); F. M. Ellis and R. B. Hallock, Rev. Sci. Instrum. **54**, 751 (1983).

[5] I. Rudnick, R. S. Kagiwada, J. C. Fraser, and E. Guyon, Phys. Rev. Lett. **20**, 430 (1968).

[6] SAES Getters/USA, Inc., Colorado Springs, CO, model FT 10+10; see also, M. Succi, R. Canino, and B. Ferrario, Vacuum **35**, 579 (1985).

[7] With the experimental chamber evacuated and surrounded by helium at 4.2 K, the current to the dispenser was ramped from 0.0 to 8.0 A at 0.1 A/min, then held constant at 8.0 A for 30 min, and then reduced to zero promptly.

[8] G. Vidali, G. Ihm, and M. W. Cole, Surf. Sci. Rep. **12**, 133 (1991).

[9] K. S. Ketola, S. Wang, and R. B. Hallock, J. Low Temp. Phys. (to be published).

[10] We use the term "wet" here in a broader sense than the usual meaning of complete wetting at saturation. The term prewetting has its usual meaning: a jump in the surface coverage followed by normal film growth.

[11] 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).

[12] D. T. Smith and R. B. Hallock, Phys. Rev. B **34**, 226 (1986).

[13] M. O. Robbins, D. Andelman, and J.-F. Joanny, Phys. Rev. A **43**, 4344 (1991).

[14] P. J. Nacher and J. Dupont-Roc, Phys. Rev. Lett. **67**, 2966 (1991).

[15] M. Schick, in *Liquids at Interfaces*, Proceedings of the Les Houches Summer School, Session XLVIII, 1988, edited by J. Charvolin, J. F. Joanny, and J. Zinn-Justin (Elsevier, New York, 1991), p. 415.