

In conclusion, a refractive-index anomaly in the region of a critical point has been observed. However, measurements are not yet sufficiently precise to specify the critical parameters that should be obtainable (i.e., ν , ξ_0 , and η) with adequate accuracy. Our observations are fitted by our theoretical relationship for the refractive index within the accuracy of our preliminary measurements.

*Work supported in part by the U. S. Atomic Energy Commission under Contract No. AT(11-1)-2203.

†Now at Middlebury College, Middlebury, Vt. 05753.

¹S. Y. Larson, R. D. Mountain, and R. Zwanzig, J.

Chem. Phys. 42, 2187 (1965).

²R. Hocken and G. Stell, Phys. Rev. A 8, 887 (1973).

³D. Bedeaux and P. Mazur, Physica (Utrecht) 67, 23 (1973).

⁴L. S. Taylor, J. Math. Phys. (N. Y.) 6, 824 (1963).

⁵R. Hocken and L. R. Wilcox, Bull. Amer. Phys. Soc. 17, 614 (1972).

⁶K. B. Lyons, R. C. Mockler, and W. J. O'Sullivan, Phys. Rev. A 10, 393 (1974).

⁷E. L. Eckfeldt and W. W. Lucasse, J. Phys. Chem. 47, 164 (1943).

⁸M. E. Fisher, J. Math. Phys. (N. Y.) 5, 944 (1964).

⁹This value of ν is referred to by J. S. Huang, W. Goldberg, and A. Bjerkaas, Phys. Rev. Lett. 32, 921 (1974). It is an unpublished result by N. C. Wong and J. S. Huang.

¹⁰P. Heller, Rep. Progr. Phys. 30, 731 (1967).

Onset Phenomena in Superfluid Helium*

T. G. Wang, M. M. Saffren, E. E. Olli, and D. D. Elleman

Jet Propulsion Laboratory, California Institute of Technology, Pasadena, California 91103

(Received 20 March 1974)

Onset phenomena in superfluid helium have been studied by measuring the rotational decay constant of a levitated, superconducting niobium sphere coated with an unsaturated superfluid film. The novel features of onset that were observed are discussed. Previous observations of a two-layer-thick solid phase of helium underlying the film have been confirmed.

Onset phenomena associated with unsaturated liquid He II film—specifically, onset of superfluidity,¹⁻⁷ superfluid density,^{8,9} and solid layers^{10,11}—have recently stimulated a great deal of interest in the scientific community. This Letter reports some novel observations of onset phenomena obtained by measuring the rotational decay time constant α of a rotating sphere coated with an unsaturated superfluid film. We believe this to be the first complete set of observations free of extensive extrapolation.

The apparatus and technique used have been described previously.¹⁰ The rotating sphere was an ultrapure, superconducting, niobium ball of 1 in. diameter and 5 μ in. sphericity. An unsaturated superfluid film in equilibrium with its own vapor at a pressure below the saturated vapor pressure was formed on the sphere's surface. The film thickness¹² is given by

$$d = [(RT/\Gamma M) \ln(P_0/P)]^{-1/3}, \quad (1)$$

where Γ is a constant,¹⁰ M is the molecular weight of He, R is Boltzmann's gas constant, T is the temperature of the sphere, P is the pres-

sure of the vapor in equilibrium with the film, and P_0 is the saturated vapor pressure. The angular velocity ω of the sphere was always kept low enough (6 to 15 rpm) that the rotational decay time constant is given by¹³

$$\alpha = -\omega^{-1} d\omega/dt. \quad (2)$$

Experimental measurements of α as a function of d were taken 1 h after the temperature and pressure had stabilized. In each set of measurements, we started with a saturated film and then slowly reduced the film thickness by removing He gas from the chamber. The results for six sets of measurements in the temperature range 1.165 to 2.08 K are summarized in Fig. 1. The points are experimental values; the solid lines are best-fit curves and do not represent any theory at this time.

We chose to analyze the data by assuming two major components in each measured value of α : the classical component that we have reported previously¹⁰ and the superfluid component unique to He II films. Figure 2 shows the experimental curve for 1.91 K (from Fig. 1) and the two com-

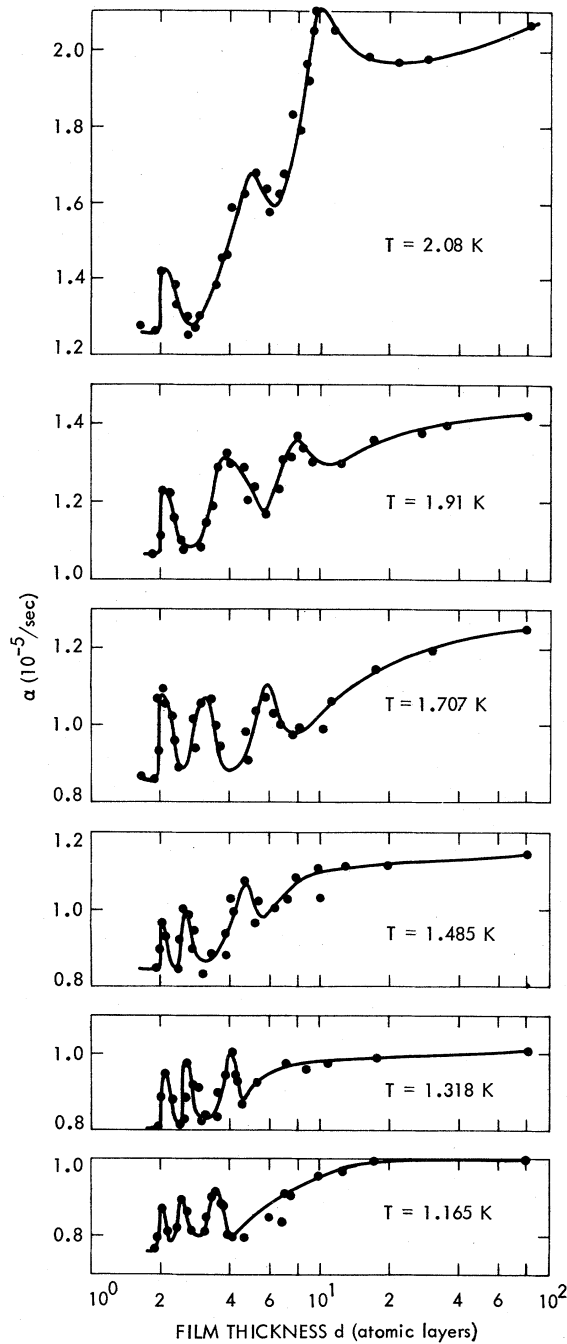


FIG. 1. Rotational decay time constant as a function of film thickness at six different temperatures. Points are experimental values. Solid lines are best-fit curves.

ponent curves, which were computer-generated from the relation $\alpha_{total} = \alpha_{superfluid} + \alpha_{classical}$. The classical component was assumed to be a smoothly varying function. The important features in this figure, to be discussed below, are

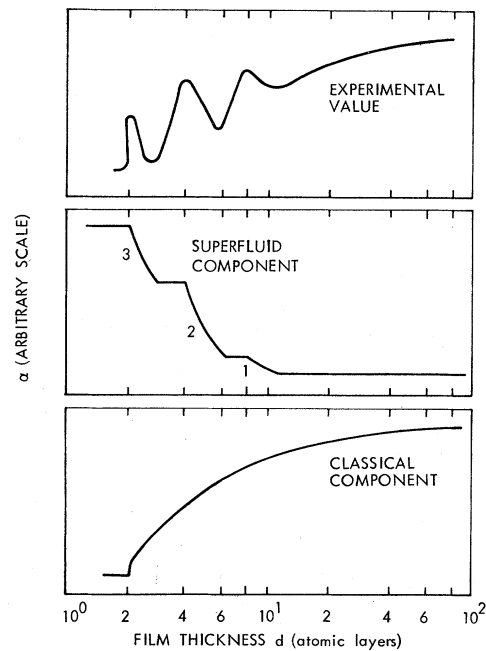


FIG. 2. Data analyzed by assuming two major contributions in our experimental value of the rotational decay time constant: a classical component and a superfluid component (example: $T = 1.91$ K). The two component curves are computer generated.

common to the data at the other temperature values.¹⁴

The first sharp rise (labeled 1) of the superfluid component is at the film thickness at which onset of superfluidity occurs.¹⁻⁷ A great deal of experimental knowledge has been obtained about this onset phenomenon. Presently, two theories concerning the mechanism of onset of superfluidity are generally favored: intrinsic fluctuation^{15,16} and the ripplon model.¹⁷ While our observations cannot be used for a definite determination of which is the proper view, they seem to favor the ripplon model. The reason for this is that ripples generated at the free surface by impinging gas molecules transfer some of the angular momentum of the impinging gas molecules to escaping gas molecules. This results in an incomplete momentum transfer between the gas and the liquid film. However, as the film thickness approaches the value at onset of superfluidity, the lifetime of ripples decreases exponentially¹⁸ resulting in a more complete transfer of momentum between gas and liquid; therefore, the free surface becomes an exponentially increasing momentum sink for impinging gas molecules. This suggests an exponential component in the rotational decay

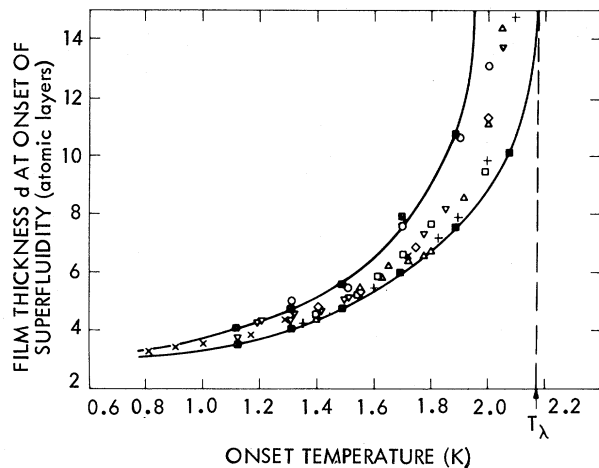


FIG. 3. Film thickness at beginning and end of onset of superfluidity as a function of temperature. The solid squares are from Fig. 1, triangles from Ref. 1, circles from Ref. 2, open squares from Ref. 3, diamonds from Ref. 4, \times -crosses from Ref. 5, plus-crosses from Ref. 7 with adjusted solid layers, and inverted triangles from Ref. 8.

rate, which we observed.

We believe that our observations are unique in that they show the beginning and end of the onset of superfluidity, as well as the onset itself. Film thickness at the beginning and end of onset is plotted as a function of temperature in Fig. 3. The square solid points are experimental values from Fig. 1, and the solid lines are best-fit curves. If these curves are indeed the boundaries of onset, then the measured values of onset of superfluidity previously reported in the literature¹⁻⁷ should lie between these curves. These values are plotted in Fig. 3 and they confirm our premise. Their positions seem to depend on the sensitivity, accuracy, and parameters of the respective experiments.¹⁹

The second sharp rise of the superfluid component is at the film thickness at which onset of superfluid density occurs. To see this, one can imagine that, although most gas molecules lose their excess momentum at the free surface, some must still be able to penetrate the first few film layers, completing the momentum transfer with the normal-fluid component in the bulk of the film. Hence, a sudden rise in the value of the rotational decay time constant is expected when the film thickness is reduced to the value for the onset of superfluid density. This film thickness has been interpreted in terms of the

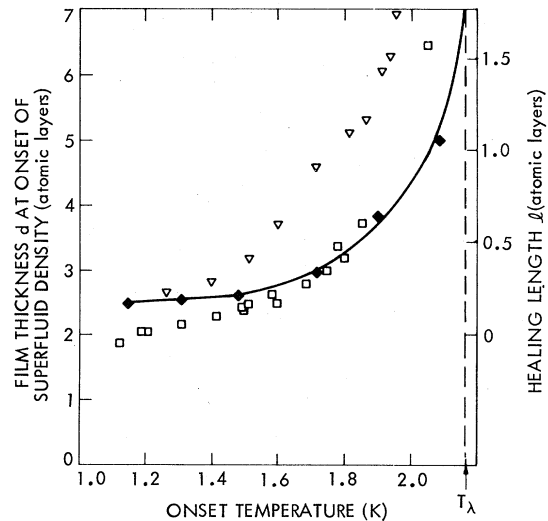


FIG. 4. Experimental values of film thickness at onset of superfluid density (left ordinate) or l (right ordinate) versus temperature. The diamonds are from Fig. 1, squares from third-sound results, Ref. 19, and inverted triangles from Ref. 21. The solid curve is from the empirical formula $d = A + BT\rho/T_\lambda\rho_s$.

Ginzburg-Pitaevskii description²⁰ of superfluid density; and the value of the superfluid healing length has been determined from it. All prior experimental data on film thickness at onset of superfluid density are extrapolated values. Our observations are presented as a function of temperature in Fig. 4 (left ordinate), with no adjustable parameters. The closed points are our experimental values, and the solid line is fitted by an empirical law of the form

$$d = A + B(T/T_\lambda)\rho/\rho_s, \quad (4)$$

where d is the film thickness, A and B are constants, T_λ is the He II transition temperature, ρ is the total density, and ρ_s is the superfluid density. The curve shown has $A = 2.0$ and $B = 0.96$. The solution of the Ginzburg-Pitaevskii equation with our proposed boundary conditions gives

$$d = d_s + 2\sqrt{2}l, \quad (5)$$

where d_s is the thickness of the solid layer and l is the superfluid healing length. Our data indicate that $d_s = 2$ atomic layers, which leads to the value $l = 0$ as T approaches 0 K (see Fig. 4). This, of course, satisfies the formula proposed by Rudnick and Fraser⁸,

$$l = \frac{\text{const } T}{\rho_s/\rho T_\lambda}. \quad (6)$$

Experimental values of l as a function of temperature are plotted in Fig. 4 using the right ordinate. The open square points are from third-sound measurements²² and the triangular points are from persistent-current measurements.²¹

The reason for the third sharp rise of the superfluid component is unknown at present. However, other authors⁹ have also observed some unexplainable feature in the region prior to the appearance of solid layers.

The general behavior of the classical component in Fig. 2 has been described before.¹⁰ We only wish to stress again that the sudden drop in value at two atomic layers is consistent with our interpretation of a solid phase two layers thick. Recently, other authors¹¹ have also observed the second solid layer in their heat-capacity measurements.

We would like to thank Ms. Hilga Campbell of the University of Southern California for her excellent help during the course of the experiment. We would also like to thank Mr. A. Barkus of the Jet Propulsion Laboratory for his editing of the manuscript. One of the authors (T.W.) wishes to thank Professor I. Rudnick and Professor F. Busse of the University of California at Los Angeles for their valuable suggestions.

*This paper presents the results of one phase of research carried out at the Jet Propulsion Laboratory, California Institute of Technology, under Contract No. NAS7-100, sponsored by the National Aeronautics and Space Administration.

¹E. Long and L. Meyer, Phys. Rev. **85**, 1030 (1952).

²E. Long and L. Meyer, Phys. Rev. **98**, 1616 (1955).

³D. J. Brewer and K. Mendelssohn, Proc. Roy. Soc., Ser. A **60**, 1 (1961).

⁴R. P. Henkel, G. Kukich, and J. D. Reppy, in *Proceedings of the Eleventh International Conference on Low Temperature Physics, St. Andrews, Scotland, 1968*, edited by J. F. Allen, D. M. Finlayson, and D. M. McCall (St. Andrews Univ. Press, St. Andrews, Scotland, 1969).

⁵K. Fokkens, W. K. Taconis, and R. de Bruyn Ouboter, Physica (Utrecht) **32**, 2129 (1966).

⁶R. Kagiwada, J. C. Fraser, I. Rudnick, and D. Bergman, Phys. Rev. Lett. **22**, 339 (1969).

⁷E. S. Sabisky and C. H. Anderson, Phys. Rev. Lett. **30**, 1120 (1973).

⁸I. Rudnick and J. C. Fraser, J. Low Temp. Phys. **3**, 225 (1970); J. C. Fraser, Ph.D. thesis, University of

California at Los Angeles, 1969 (unpublished).

⁹M. Chester and L. C. Yang, Phys. Rev. Lett. **31**, 23, 1377 (1973).

¹⁰T. G. Wang, Phys. Rev. Lett. **31**, 6, 344 (1973).

¹¹M. Bretz, Phys. Rev. Lett. **31**, 24, 1447 (1973).

¹²At this time, we assume that the density of the film is uniform throughout and that the entropy of the adsorbed film is the same as that of the bulk liquid.

¹³The equation for calculation of accommodation coefficient from rotation decay measurements in Ref. 10 is incorrect. Theoretical and experimental efforts are underway to redefine the correlation.

¹⁴The separation of the experimental curve into two components confirms our belief that the classical treatment of vapor-liquid interaction which ignores the quantum nature of the superfluid-helium surface cannot be applied rigorously to the superfluid-helium system [see M. Cole, Phys. Rev. Lett. **28**, 1622 (1972)]. The vapor-liquid interface of superfluid helium differs significantly from that of a classical liquid; for one thing, it has to serve as a bridge between the Maxwellian and the boson systems. Based on the above observations, one should not expect any significant change in the rotational decay time constant from the interface region all the way to the bulk region. This leads to the conclusion that the accommodation coefficient at the film interface cannot be too different from the bulk value, measured by K. R. Atkins, B. Rosenbaum, and H. Seki [Phys. Rev. **113**, 751 (1959)] and G. H. Hunter and D. V. Osborne [J. Phys. C: Proc. Phys. Soc., London **2**, 2414 (1969)]. However, we cannot prove this at this time.

¹⁵S. V. Jordanskii, Zh. Eksp. Teor. Fiz. **48**, 708 (1965) [Sov. Phys. JETP **21**, 467 (1965)].

¹⁶J. S. Langer and M. E. Fischer, Phys. Rev. Lett. **19**, 560 (1967).

¹⁷C. G. Kuper, in *Superfluid Helium*, edited by J. F. Allen (Academic, New York, 1966).

¹⁸The ripplons become third sound in the thin film, and there is experimental evidence [see T. G. Wang and I. Rudnick, J. Low Temp. Phys. **9**, 425 (1972)] which shows that the third sound attenuates exponentially as film thickness approaches the value at onset of superfluidity.

¹⁹It is worth pointing out here that although ρ_s at the surface is being depleted, the ρ_s average of the system does not have to change significantly [see D. L. Goodstein and R. L. Elgin, Phys. Rev. Lett. **22**, 383 (1969)].

²⁰V. L. Ginzburg and L. P. Pitaevskii, Zh. Eksp. Teor. Fiz. **34**, 1240 (1958) [Sov. Phys. JETP **7**, 858 (1958)].

²¹R. P. Henkel, E. N. Smith, and J. D. Reppy, Phys. Rev. Lett. **23**, 1276 (1969).

²²For a recent paper on the subject, see J. H. Scholtz, E. O. McLean, and I. Rudnick, Phys. Rev. Lett. **30**, 4 (1974).