perimental errors are large enough to permit this behavior. As a third alternative, however, we recognize that there are appreciable ϵ-dependent contributions to u_2 which do not affect the asymptotic behavior, and it is not clear whether these are considered correctly by dynamic scaling. If the prediction Eq. (2) pertains only to the asymptotically dominant terms, one might use $u_2 \propto \epsilon^{0.33}$ and $\xi \propto \epsilon^{-0.67}$ for all X and predict a concentration-independent $z \simeq 0.34$. This value falls within the permitted range of the experimental result. Nonetheless, we remark that the measured z for $X = 0.1$ tends to be higher than any of the predicted values. There has been a tendency also for measurements[15,16] of other transport properties to yield exponents which are larger than the predictions.

One might expect also to obtain the approximate amplitude of D from dynamic scaling by setting numerical constants equal to unity and treating the asymptotic proportionality $D_A \sim u_2\xi$ as an equality. However, the large latitude in the choice for ξ and u_2, and the unknown size of Z_2 in Eq. (1), permit at best an estimate for the order of magnitude of D. For $X = 0$ and at $\epsilon = 10^{-4}$ one obtains $D \simeq u_2\xi \simeq 1.2\times10^{-3}\ \mathrm{cm}^2/\mathrm{sec}$ if one uses $\xi = \xi_0\epsilon^{-0.67}$, $\xi_0 = 2\times10^{-8}$ cm, and the measured u_2.[12] With a characteristic length for our geometry of 0.7 cm, this yields $\tau \simeq 400$ sec at $\epsilon = 10^{-4}$ and $X = 0$. This is consistent with the experimental values of $\tau(X)$ quoted above and shown in Fig. 3.

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Third Sound and the Healing Length of He II in Films as Thin as 2.1 Atomic Layers*

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Measurements of the velocity of third sound on films as thin as 2.1 atomic layers yield the healing length of superfluid He II down to temperatures of 0.1 K. It is argued that these films are two-dimensional superfluids.

The role of dimensionality in determining the properties of ordered phenomena such as magnetism, superconductivity, and superfluidity is a subject of continued and current interest. Of particular concern is the question of whether coherent phenomena observed in three-dimensional configuration space persist in two-dimensional space.[1] He II is distinguished by the occurrence of extremely thin uniform films (measured in angstroms or atomic layers) and it is fair to regard them as two-dimensional if such a concept has any meaning for actual systems. There is a growing body of experimental knowledge[2] about their superfluid properties, and, especially with the results reported here, the existence of superfluidity in two-dimensional systems is well established. We report here a systematic investigation of superfluidity in films as thin as 2.1 atomic layers total thickness in the temperature range 0.1 to 1.2 K through the use of third sound. The measurements yield the quantity from which the healing length is determined. Taken together with previous measurements at higher temperatures, this quantity is now known at all temperatures. Although these films are very thin, we find no obvious breakdown in the thermohydrodynamics which is the basis for the description of third sound (unless the necessity of introducing the notion of a healing length is considered evidence of such a breakdown). It is remarkable that in He II, quantum mechanics operates on a macroscopic scale; thermodynamics and hydrodynamics on a microscopic scale.

When a film of He II is disturbed, surface waves, called third sound, spread out from the disturbance much like ripples on a water surface. These waves are characterized by the fact that the normal component is clamped by viscosity, and only the superfluid component moves. The wave velocity C_3 is given by[3]

$$C_3^2 = (\langle \rho_s \rangle / \rho) f d, \tag{1}$$

where $\langle \rho_s \rangle / \rho$ is the superfluid fraction of the film averaged over the thickness d, and f is the Van der Waals force of attraction between the helium and the substrate. This force is the derivative of the chemical potential μ, $f = d\mu/dy|_{y=d}$, where μ is given by

$$\mu = -\alpha / y^3. \tag{2}$$

The quantity α is a constant characteristic of the substrate.[4] The existence of third sound is evidence of superfluidity, and a measurement of C_3 can be used to determine $\langle \rho_s \rangle / \rho$ when α is known. The thickness d of unsaturated films can be determined by measuring the pressure p of the gas in equilibrium with the film at a temperature T. Since the chemical potential of the film and vapor are equal,

$$\alpha / d^3 = (kT/m) \ln(p_0/p), \tag{3}$$

where p_0 is the saturated vapor pressure, k is the Boltzmann constant, and m is the mass of a helium atom.

For the thinnest films used here, the Mach number, $M = C_3/C_1$ (where C_1 is the velocity of first sound), is not small and the incompressibility condition used in obtaining Eq. (1) is not justified. Under these conditions the velocity of third sound is given by

$$C_3^2 = (\langle \rho_s \rangle / \rho) f d [1 + (\rho / \langle \rho_s \rangle) M^2]^{-1}. \tag{4}$$

In the thinnest films this leads to an 8% correction of C_3. The third-sound measurements are made in the conventional way,[3] by measuring the time of flight of a pulse. The test chamber has in it a film reservoir composed of about 6 g of 500-Å aluminum-oxide powder to give a surface area of about 500 m^2. The free volume is kept small so that below 1.5 K practically all the helium is in the film, and thickness changes caused by vapor condensation as the temperature is lowered are very small. The temperature range studied is from 1.2 down to 0.1 K. For temperatures down to 0.5 K, a helium-3 cryostat is used. The film is condensed at 1.5 K and its thickness is determined by measuring T and p and using Eq. (3). For temperatures down to 0.1 K, a dilution refrigerator is used and the film thickness is determined by measuring the value of C_3 at temperatures higher than 0.5 K, where the dependence of C_3 on film thickness had already been established. We have made measurements on a polished CaF_2 single-crystal substrate and on a glass substrate. Equation (3) is often written in the form $d^3 = \Gamma[T \ln(p_0/p)]^{-1}$, where d is now in atomic layers. The values of Γ used in determining the thickness of the films were 23 $(\text{layers})^3$ K for CaF_2 and 27 $(\text{layers})^3$ K for glass, in agreement with the values given by Sabisky and Anderson.[2,4] As a result of their experiments, the Van der Waals constants are now accurately known and considerable confidence can be placed in the values of film thickness. Figure 1 shows some of the experimental results.

From measurements of C_3, $\langle \rho_s \rangle / \rho_{sb}$ can be determined from Eq. (4). (ρ_{sb} is the bulk superfluid density.) Our data provide confirmation[5,6] that the depletion in $\langle \rho_s \rangle$ follows the law

$$\langle \rho_s \rangle / \rho_{sb} = 1 - D/d \tag{5}$$

for small thickness d, as shown in Fig. 2. D is a temperature-dependent constant equal to the intercept of the line in Fig. 2. In determining D, the line is always drawn with a 45° slope and so that it best fits the small-thickness data as shown in Fig. 2.

At present there is no single accepted view of the way in which ρ_s is depleted. The layer of helium next to the substrate is under high pressure

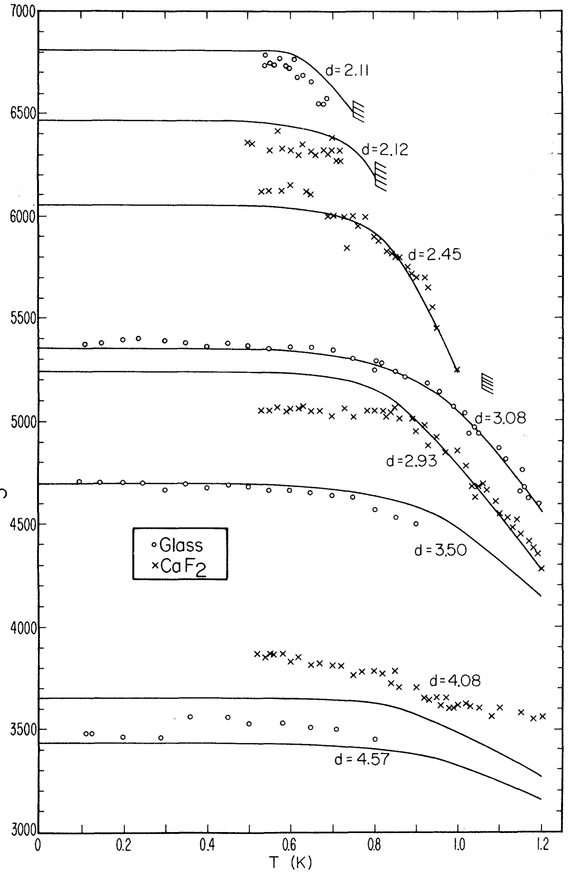

FIG. 1. Third-sound velocity C_3 versus temperature for a variety of thicknesses on both glass and CaF$_2$. The thicknesses of the films in atomic layers are recorded on the graph. The solid lines are calculated from Eqs. (4) and (5) using the values of D shown in Fig. 3. The hatched areas show the onset temperature for the three thinnest films.

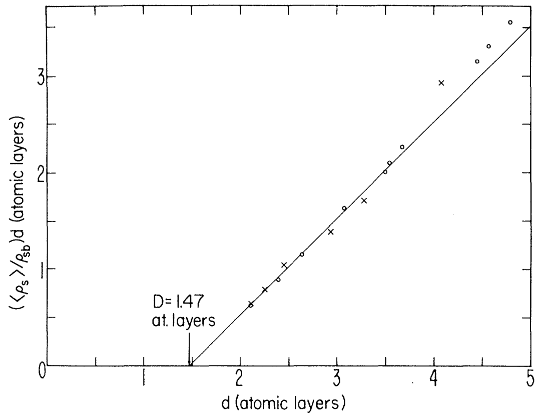

FIG. 2. Plot of $\langle\rho_s\rangle/\rho_{sb}$ versus d for helium films on glass (circles) and CaF$_2$ (crosses) at 0.5 K. The quantity $\langle\rho_s\rangle/\rho_{sb}$ is obtained from third-sound-velocity measurements. The film thickness d comes from measuring the chemical potential of the film at 1.5 K. The intercept of the line with the abscissa gives D, the effective nonsuperfluid thickness of a helium film at 0.5 K.

because of Van der Waals forces, and it is reasonable to assume that in a thickness d_s the helium is solid because this pressure exceeds the melting pressure. Chan *et al.* report evidence that replacing the first layer of helium by one of neon leaves the superflow properties of the film unchanged.[7] In the liquid one can argue that ρ_s must satisfy a condition that it vanish at the boundaries, achieving its bulk value only in the interior of thick films. If this occurs equally at the solid layer and the free surface of film, then

$$D=d_s+2a, \tag{6}$$

where a is called the superfluid healing length.[8] An alternative point of view is that ρ_s vanishes at the solid boundary and satisfies some mixed boundary condition at the free surface. Sobyanin has advanced arguments in favor of the condition that $\rho_s=0$ at the surface.[9] It is also possible that when ρ_s is depleted, the depletion occurs more or less uniformly throughout the thickness of the film. Our experimental results cannot be used to determine which point of view is correct. To carry the discussion further we shall discuss the results in terms of Eq. (6) with $d_s=1$. Figure 3 is a plot of D, or a, as a function of temperature, determined by the kind of data shown in Fig. 2. The results of other investigations on films at higher temperatures are also shown.[10] Several points can be made: Our results are consistent with previous third-sound results and those obtained by a quartz-microbalance technique by Chester and Yang,[11] and appear to be somewhat lower than the results of perisitent-current measurements.[5] It is worth emphasizing that there are no adjustable constants in the third-sound measurements, and that these measurements now give D and the healing length from 0.1 to 2.05 K. Above about 1 K the curve can be fit by an empirical law of the form $D=A+B(T/T_\lambda)\rho/\rho_{sb}$, where A and B are constants. The curve shown has $A=0.5$ and $B=1.13$. Between 0.1 and 0.6 K, D has the constant value of 1.47 atomic layers with a standard deviation of 0.01 atomic layers. If $d_s=1$ atomic layer, then $a=0.235$ atomic layers or 0.846 Å as $T\to 0$. This compares with the value 1.00 ± 0.10 Å for the hollow-core radius of a quantized vortex ring.[12] Henkel[13] from fourth-sound data reports values of D of 0.7, 1.0, 1.5,

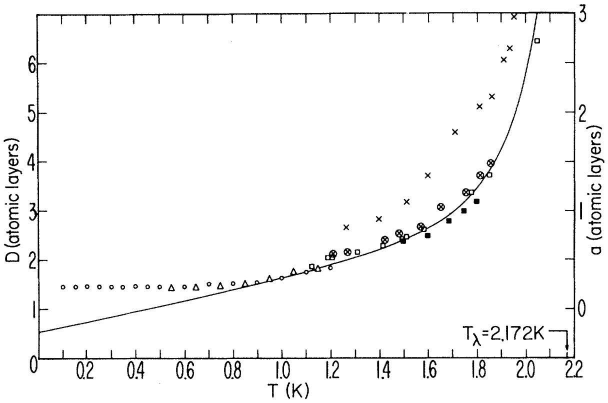

FIG. 3. Graph of the experimental values of D (left ordinate) or a (right ordinate) versus temperature. The circles and triangles are the present results on glass and CaF_2, respectively. The open squares are previous third-sound results at higher temperatures (Refs. 3, 6, and 10), and the crosses are persistent-current results (Ref. 5). The filled squares are unpublished results obtained in this laboratory by K. L. Telschow. The circled crosses are the results of a quartz-microbalance experiment by Chester and Yang (Ref. 11). The solid curve is from the empirical formula $D=A+B(T/T_\lambda)\rho/\rho_{sb}$.

or 2.0 atomic layers at $T=0$, depending on the way in which the data are normalized. If $d_s>1$ atomic layer, our data yield very small or vanishing healing lengths as $T\rightarrow 0$. From the Gross-Ginzberg-Pitaevskii theory one concludes that the surface tension is proportional to $\frac{1}{2}\rho_s\hbar^2/m^2a$. Our values of a are consistent with the known value of the surface tension. Radically smaller values of a, as would be obtained for $d_s\rightarrow 1.47$, are inadmissible if this expression is correct. If, in fact, the zero-degree healing length a controls the surface tension, then the requirement of the third law of thermodynamics[14] that the surface tension is temperature independent at $T=0$ also constrains a to be temperature independent, as observed.

We have benefitted greatly from the comments of Seth Putterman who derived Eq. (4) and pointed out to us the connection between the surface tension and the healing length as $T\rightarrow 0$. Peter Seligman is largely responsible for the construction of the dilution refrigerator. W. Gilbert Clark provided the facilities of his laboratory in the use of the refrigerator. Francis Forrette and Dave Scholler gave invaluable assistance during the measurements and in analyzing the data.

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[10] In *all* previous publications from this laboratory, film thicknesses on glass have been calculated using a value of $\Gamma=87$ (layers)^3 K. There had been mounting evidence that this value (obtained on argon-coated copper) was inappropriate for glass but we have resisted using different, tentative values for Γ. [This situation is mentioned in T. Wang and I. Rudnick, J. Low Temp. Phys. 9, 425 (1972).] We believe that the evidence now for a value of $\Gamma=27$ is convincing and *all* previous film thicknesses reported by us can be corrected by multiplying by the factor $(\frac{27}{87})^{1/3}=0.677$. This has been done for the data in Ref. 7 which appear in Fig. 3. For a discussion specifically directed at the evidence supporting the use of this lower value of Γ for glass rather than 87, see Ref. (4), bottom of page 801 and page 805, and Ref. (2).

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Next-to-Leading Terms in the Correlation Function inside the Scaling Regime

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An application of Symanzik's method for treating exceptional momenta (the Callan-Symanzik equations combined with operator-product expansions) yields the leading corrections to the large-momentum behavior of the correlation function near the critical point. The result is used to argue that the conclusion drawn from numerical calculations, namely, that strong scaling is violated, is unwarranted.

The leading behavior of the renormalized inverse spin-spin correlation function[1] $G^{-1}(p,\xi)$ for $p\xi \gg 1$ and $pa \ll 1$, where ξ is the correlation length and a is the lattice spacing, has been calculated by a few methods. The result is $G^{-1} \propto \xi^{-\eta} p^{2-\eta}$,[2,3] where the exponent η was given as an expansion in $\epsilon = 4-d$,[4] d being the number of dimensions, or in $1/n$,[5] n being the number of components of the order parameter. The subsequent terms in $(p\xi)^{-1}$ in the scaling region have attracted considerable attention, being relevant both to the interpretation of neutron-scattering experiments[6] and to the theoretical discussion of the connection of the spin-spin correlation function to the specific heat (or the energy-energy correlations)[7] and to the susceptibility.[8] Furthermore, they are connected to the wider question of whether strong scaling holds in three-dimensional systems.[9] In order to determine what kinds of powers are present beyond the leading one, one can perform, say, an ϵ expansion of all the diagrams, keeping track of their nonleading behavior, provided one knows in advance what and how many powers are present. This work is devoted to that question. For that purpose, we have used the Callan-Symanzik (CS) equations as in Ref. 3. The advantage is that they produce directly the power behaviors, rather than a series of logarithms, which have to be exponentiated. As we shall see below, the correction is a combination of two competing powers, which makes the CS formalism particularly suitable. The problem is that one has to go beyond the study of homogeneous CS equations[3] and consider the right-hand sides (rhs) of these equations in order to obtain the corrections to the main power. This leads to the problem of determining the asymptotic behavior of correlation functions in which some subsets of momenta remain finite. This problem can be solved with the use of the Wilson-Kadanoff[10] short-distance expansion (SDE) of an operator product. Its relevance to this problem has been first discussed in field theory by Symanzik,[11] and our discussion will follow closely his analysis of the exceptional momenta. The result is

$$G^{-1}(p,\xi) \underset{p\xi\gg 1}{\sim} \xi^{-2}(p\xi)^{2-\eta}\left[A + B(p\xi)^{-1/\nu} + C(p\xi)^{-(1-\alpha_s)/\nu}\right], \tag{1}$$

where A, B, and C are constants whose ϵ or $1/n$ expansion may be computed in perturbation theory. The terms in $(p\xi)^{-1}$ which are neglected are governed by powers close to 4 in four dimensions, whereas $1/\nu$ or $(1-\alpha_s)/\nu = 2 + O(\epsilon)$. If we express ξ in terms of the reduced temperature $t = (T - T_c)$ by $\xi = \xi_0 t^{-\nu}$, and go to the bare correlation function, Eq. (1) becomes

$$G_0^{-1}(p,t) \sim (p\xi_0)^{2-\eta}\left[A + Bt(p\xi_0)^{-1/\nu} + Ct^{-(1-\alpha_s)}(p\xi_0)^{-(1-\alpha_s)/\nu}\right]. \tag{2}$$

This result has only been derived for T larger than T_c. The C term has been predicted already as necessary for reproducing the specific-heat anomaly from the behavior of the correlation function.[7]