Superfluid Onset, Frequency Response, and Connectivity*

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A recent study of thin He^4 films by Chester, Yang, and Stephens using a quartz-microbalance technique has shown frequency anomalies ascribed to the onset of superfluidity. The data imply that superfluid is present at thicknesses less than those at the anomalies. We propose a model involving surface heterogeneities to explain the results. Using the same model, it is possible to resolve several puzzles associated with other studies of onset, including the anomalous attenuation of third sound.

I. INTRODUCTION

Chester, Yang, and Stephens[1] have recently studied the onset of superfluidity in thin He^4 films using a high-frequency quartz microbalence. This technique relies upon the change in shear-mode resonant frequency of a crystal owing to mass loading when a thin film is absorbed on its surface. The method has been applied to adsorption studies of heavier gases[2] but had not been used previously for helium. Its applicability to studies of superfluid onset depends upon the absence of viscosity in the superfluid fraction; therefore, one might expect that the frequency shift caused by a He^4 film at temperatures T below the superfluid-onset temperature T_0 will be less than for $T > T_0$. Indeed, the recent experiments disclose anomalies in the frequency–vapor-pressure isotherms which are closely correlated with T_0 values obtained by other methods. However, the new results show onset occurring at somewhat lower pressures and smaller film thicknesses.

Although the quartz-microbalance anomalies are evidently associated with superfluidity, we disagree with the interpretation of Chester *et al.* and we here propose an alternative explanation. In preface we remark that, rather than implying any criticism of their work, we believe that their results have contributed an important clue to the understanding of earlier studies of onset in thin films.

II. FREQUENCY RESPONSE OF UNIFORM FILMS

We first consider the quartz microbalance in terms of an idealized model; an infinite-plane solid surface bearing a liquid layer of uniform thickness. If the solid oscillates at some frequency ω in a transverse direction, a portion of the fluid will be carried along owing to its viscosity. For an ordinary fluid of viscosity η and mass density ρ, the quantity of liquid moving with the surface is approximately that portion contained within a boundary layer of thickness $(\eta/\omega\rho)^{1/2}$. If the liquid is effectively composed of normal and superfluid fractions ρ_n and ρ_s with viscosities η_n finite and $\eta_s = 0$, then the boundary-layer thickness is determined by ρ_n and η_n. For film thicknesses much smaller than $(\eta_n/\omega\rho_n)^{1/2}$, all of the normal fluid in the film will be effectively locked to the surface and will contribute to the inertial mass of the crystal.

We now consider the situation for a finite crystal. In this case both normal and superfluid fractions at the forward and rear edges are constrained to move with the surface. For frequencies $\omega \to 0$, the boundary conditions cause even the superfluid to be effectively locked to the surface. But for very high frequencies, the boundary motion can only be transmitted to the superfluid in a narrow region near the edges, that within a distance of the order of a wavelength of surface waves. Since the normal fluid is everywhere locked to the surface by its viscosity and it is only the relative motion of superfluid that is of concern, the boundary motion is communicated to the superfluid via third sound.[3] Thus the quantity of superfluid moving with the crystal at high frequencies is proportional to u_3/ω, where u_3 is the speed of third sound. If the characteristic dimension of the crystal surface is L, the condition for slippage of an appreciable part of the superfluid is $\mu_3/\omega \lesssim L$. Applying this criterion to the experiments of Chester *et al.*, where $\omega = 24$ MHz, $L = 3$ mm, with experimental u_3 values appropriate to the thicknesses and temperatures of the films in the microbalance study, one finds that the condition for slippage of a major portion of the superfluid is well satisfied over the full range of the experiments.

Chester *et al.* did not present an analysis of the effect of boundary motion, but their interpretation of the experiment seems consistent with the above discussion. However, we believe that the actual conditions near onset are considerably more complicated and this is indicated by a closer inspection of the data.

In Fig. 1 we see that the designated onset points mark local maxima in the frequency–vapor-pressure isotherms: For a region of pressures beyond "onset," the resonant frequency decreases with increasing pressure before turning up again. The ef-

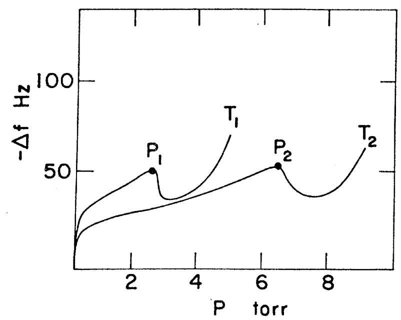

FIG. 1. Schematic data of Chester, Wang, and Stephens (Ref. 1) showing typical frequency-response isotherms of the quartz-crystal microbalance vs He^4-gas pressure. The points labeled P_1 and P_2 are designated as the vapor pressures of superfluid onset at T_1 and T_2 ($T_2 > T_1$).

fect is most pronounced at low temperatures and is not present in He^3 films; therefore, it seems clear that the anomalies are assoicated with superfluidity. However, if we interpret the results according to the preceeding analysis, it appears that, in the region just beyond the maximum in the curve, the total quantity of normal fluid in the film is decreasing as more film is added. Thus, if a film at onset is entirely normal, it seems that the addition of more helium causes a significant portion of the original film to convert to superfluid. This implies that, contrary to the conventional microscopic theory[4–6] the healing length of the superfluid order parameter at the solid boundary is strongly reduced as the free-surface boundary recedes. One possible resolution to the puzzle is to assume that the film at onset already contains an appreciable quantity of "superfluid" which is somehow unable to flow until the thickness is increased. Experimental evidence for superfluidity without superflow has been obtained previously from third-sound experiments.[3] The recent microbalance experiments have brought the problem into sharper focus and lead us to suggest a mechanism for the phenomenon.

III. CONNECTIVITY OF HETERGENEOUS FILMS

We believe that the actual films are heterogeneous, i.e., with appreciable variations in substrate binding energy from point to point on the surface. It is well known that typical surfaces have large variations in binding due to faceting, strains, dislocations, and impurities.[7] If the heterogeneity has an appreciable long-range component[8] an adsorbed film will form regions of varying thickness, i.e., "puddles" connected by regions of thinner film. Any superfluid component in a puddle of lateral dimension l will be locked to the substrate if $u_3/\omega >> l$ and, hence, will appear as normal fluid. But as the vapor pressure is increased, a point will be reached at which the film in the bridging regions between puddles becomes thick enough to "conduct" superfluid between adjacent puddles causing an increase in the effective dimension from l to $\sim L$. When this occurs, superfluid, which had been present before "onset" but locked to the substrate, is freed and the inertial mass of the film is reduced. For actual surfaces there must be a distribution of bridging-film thicknesses, puddle sizes, and separations, so that the freeing process occurs gradually over a range of pressure. On this basis, we estimate that the characteristic size of the puddles is on the order of 10^{-5} cm or less.

The region of decreasing frequency beyond the maxima are here interpreted in terms of the increasing connectivity of the film, where the effective dimension is increasing from l to L. Since other methods for detecting onset require connectivity on the order of a few centimeters, it is more appropriate to compare the microbalance data with other results by selecting as "onset" points the regions of the frequency minima where the response returns to a positive slope. On this basis, the microbalance results agree with older work, lying on the common curve of onset pressure versus temperature[3] to within experimental scatter.

IV. OTHER EXPERIMENTS

Our model offers some resolution to persistent difficulties in understanding other onset experiments. In the experiments of Rudnick and collaborators,[3,9] it is found that third-sound signals are strongly attenuated as the film thickness is reduced, whereas u_3 is hardly affected, indicating the vanishing of superflow at a finite ρ_s/ρ. The origin of the strong attenuation is a matter of current dispute,[10–14] but all theories share the common feature of assuming that the dissipative mechanisms are intrinsic properties of films on uniform substrates. However, it is highly likely that the substrates used in these experiments have typical heterogeneities and, therefore, tend to form separated puddles when the pressure is reduced. Since the third-sound transmitter and receiver are separated by a fixed distance on the order of a few centimeters, the transmission of signals requires that there be a continuous conducting path bridging many puddles at "onset." In the region of onset there will be an irregular network of paths whose connectivity changes rapidly with the pressure. The changing connectivity will appear as an overall impedance while the transmission speed remains approximately constant.

This general picture also seems applicable to the recent persistent-current experiments of Chan, Yanoff, Pobell, and Reppy.[15] In these studies, it was found that a small increase in film thickness caused an abrupt appearance of a persistent current. This jump in persistent angular momentum corresponds to an increase of superfluid mass greater than the increase in total mass of the film. Here, too, we believe that the explanation lies in the rapidly growing connectivity of the film.

Evidence that heterogeneity plays a strong role in the flow of thin He^4 films is provided by a recent study using graphite surfaces.[16] The atypical uniformity of graphite substrates is manifested by the specific-heat behavior of adsorbed He^3 and He^4 monolayers.[17–19] In the mass-flow studies using graphite, there are very large increases in flow rate which appear related to the onset points seen in films on other surfaces. On graphite, however, the increases occur at appreciably lower pressures and somewhat smaller thicknesses. The comparison indicates that the heterogeneity on typical substrates can influence the properties of He^4 films of several layers thickness.

The connectivity of the film in our model bears a strong similarity to the question of electrical conductivity of disordered semiconductors; This problem is a topic of current interest as an application of percolation theory.[20,21] Films would provide an interesting two-dimensional subject for the theory.

Finally, we suggest that the ideas proposed here can be put to an experimental test. The apparent onset of superfluidity in the microbalance method depends on the slippage of ρ_s at the frequency of the crystal. Therefore, if one compares the crystal response at two different frequencies, the anomaly should appear at a lower pressure at the higher frequency. On crystals with more uniform substrates, the anomalies should appear earlier and should also tend to disappear entirely, approaching the response of an ideal surface. Such a study is under way in this laboratory.

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Exchange Energies and Potentials at Finite Temperatures

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The exchange energy at finite temperature is discussed as a first-order perturbation. Consistent thermodynamical definitions of the average exchange potential are suggested for a Hartree–Fock–Slater calculation. It is shown that the exchange interaction satifies the virial theorem.

In a recent paper[1] a Hartree–Fock–Slater (HFS) method was suggested for calculations of the electronic levels and the equation of state for matter at finite density and temperature. It was assumed in this paper that the atomic electrons occupy single-particle levels according to Fermi statis-