# Mass Transport of He⁴ Films Adsorbed on Graphite

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Flow of He⁴ films adsorbed on graphite is qualitatively different from flow on other substrates. We observe two distinct types of "onset" phenomena. Type $A$ appears for films thicker than 2 layers at temperatures $T_A$ within 20 mK of $T_\lambda$. In thicker films there is an additional more dramatic increase (onset $B$) at lower $T$. $T_B$ varies with $P/P_0$, and is higher than temperatures of "superfluid onset" reported previously.

Thin films adsorbed on various substrates have superfluid onset temperatures $T_0$ which vary with film thickness (or relative pressure $P/P_0$) in a regular manner, so that results of a large number of studies fall on a common curve of $T_0$ versus $P/P_0$.[1-3] However, some recent experiments[4-7] using presumably more uniform surfaces have yielded somewhat higher $T_0$ values. These results raise doubts as to the fundamental nature of the common curve of onset temperatures, and suggest that in typical multilayers the range of effects due to substrate heterogeneity may extend well beyond the first layer. This conjecture is supported by our current studies of flow on graphite, which according to recent heat capacity measurements[8-10] is a manifestly more uniform surface. In this Letter we report a complex of "onset" phenomena which have not been seen in films on typical substrates.

Our study is of mass transport of He⁴ multilayers on Grafoil,[11] along directions parallel to the basal-plane graphite surfaces. The structure of Grafoil, inferred from manufacturer's information, x-ray diffraction, optical and electron microscopy, gas adsorption, and calorimetry, consists of laminas of exfoliated natural graphite, oriented and interleaved as in a loose stack of cards. Typical crystallite dimensions are 5 $\mu$m laterally between visible line defects and a few hundred angstroms in thickness. The crystallites are oriented with basal planes parallel to the Grafoil sheet surface, with mean deviation ~7 deg. Through the thickness of a 0.01-in.-thick sheet there are ~6000 basal planes exposed for gas adsorption. The material has very high chemical purity and can be heated to high temperatures to remove adsorbed gases. These features are important, we believe, in understanding the distinctions between the flow properties of He⁴ in the Grafoil flow channel and the flow of films on chemically and crystallographically less uniform substrates.

The low-temperature cell consists of two chambers connected through a Grafoil gasket, permitting surface transport along the basal-plane direction. The flow rates $R$ were measured by steadily pumping the exit chamber through a calibrated mass-spectrometer leak detector (except that the high rates below onset $B$ were estimated from the dynamic pressure rise in the pumping line). The vapor pressure $P$ in the "supply" chamber was stabilized by the presence of a quantity of Grafoil acting as ballast. $P$ and the liquid-helium bath pressure $P_0$ were monitored by a capacitance gauge. Except for the highest flow rates, the depletion of adsorbate from the supply chamber was negligible over the course of an experimental day.

Sample results of a survey of twenty coverages are shown in Figs. 1 and 2. We distinguish four distinct regimes, as follows:

(a) At lowest $P/P_0$, $R \propto \exp(-q/k_B T)$, with $q$ $\simeq$ the heat of adsorption. The exponential law is consistent with "normal" transport of film and/or vapor driven by a pressure difference equal to the vapor pressure. Pollock *et al.*[12] found similar $T$ dependence for arrival times of low-density He gas pulses traveling along tubes at low temperatures, and they showed such behavior to be consistent with migration times of adsorbing-desorbing gas.

(b) At relative pressures $P/P_0 \gtrsim 0.02$, $R$ begins to rise above the exponential law at $T \simeq 2.4$ K. The rise is smooth and increases monotonically as $T$ falls. This regime is termed the "precursor."

(c) The precursor is followed by a relatively abrupt increase ("onset $A$") at $T_A \simeq T_\lambda$. Although $T_A$ is effectively constant, the initial rate of increase and dependence at lower $T$ change markedly and nonmonotonically with $P/P_0$.

(d) There is a second and more violent rise ("onset $B$") at lower $T$. The rates first increase gradually and then rise more steeply: Increases
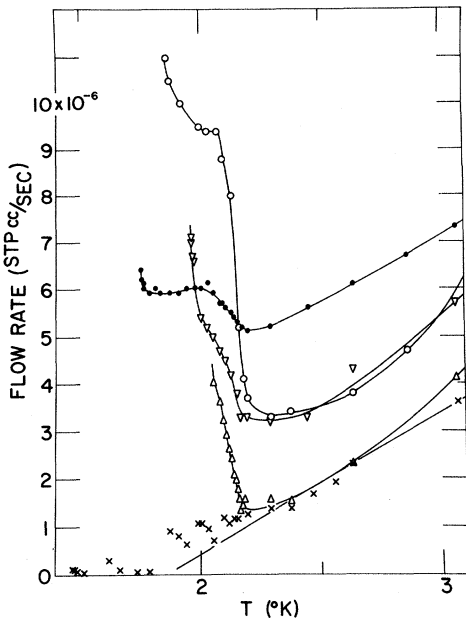
FIG. 1. Flow rates of He$^4$ for coverages exhibiting onset $A$. Coverages correspond to partial pressures $P/P_0(T_\lambda)$ as follows: crosses, $4.3 \times 10^{-4}$; triangles, 0.0179; open circles, 0.0294; inverted triangles, 0.0529; closed circles, 0.1500.



FIG. 2. Flow rates of He$^4$ for higher coverages exhibiting onset $B$. Coverages correspond to fractional pressures $P/P_0(T)$ as follows: closed circles, 0.15; plusses, 0.76; triangles, 0.80; inverted triangles, 0.87.

are typically greater than a factor $10^3$ on cooling 0.1 K below $T_B$. $T_B$ increases monotonically with $P/P_0$.

Of these observations, it is the trend of $T_B$ with $P/P_0$ that is most comparable to previous work, resembling the common curve of "super-fluid onset" temperatures $T_0$. In Fig. 3 we compare our $T_B$ values with the collected results of work on other surfaces, using several different techniques.[1-3] The $T_B$ and $T_0$ curves are generally of the same shape, but $T_B$ values are considerably higher: The shift at $P/P_0 = 0.61$ is ~0.3 K. Alternatively, our $T_B$ values occur at much lower pressures than those corresponding to the common $T_0$ curve or even of the recent shifted results[4-7]; nevertheless, the similarities in the curves of $T_B$ and $T_0$ strongly suggest that they correspond to the same basic phenomena, albeit influenced by substrate effects. Since the graphite surfaces are known to be more uniform than typical substrates,[13,14] the higher onset temperatures ($T_B$) are more representative of "ideal" films. The mechanism by which superfluidity might be suppressed by surface heterogeneity is not known, however, or if it does indeed suppress it: There even exists a theoretical model in which superfluidity is enhanced by heterogeneity.[15]

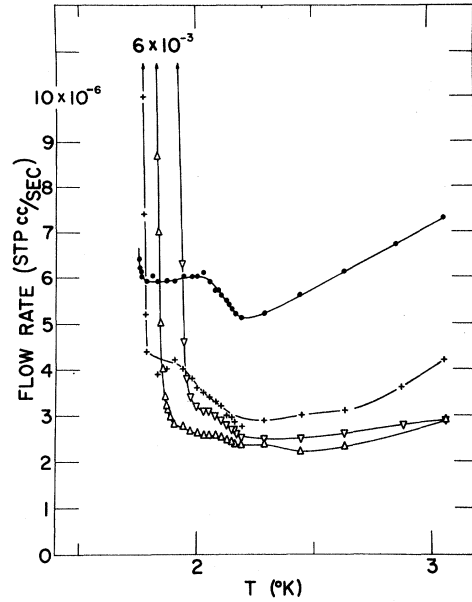The occurrence of onset $A$ is almost but not

completely novel. Long and Meyer[16] reported transitions in the flow rates of their films occurring just at $T_\lambda$. These transitions were obtained for only one experimental configuration, involving very small pressure heads. For more conventional arrangements with large pressure differences the flow remained normal down to temperatures near the common $T_0$ curve. Long and Meyer could not explain the higher temperature transitions nor repeat them with other arrangements; neither have they been duplicated by others. Our type-$A$ onsets may be basically the
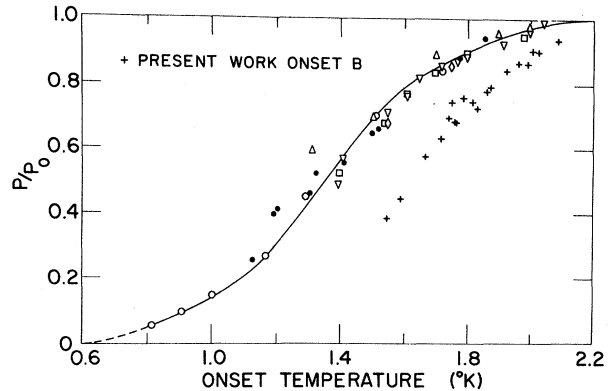


FIG. 3. Onset curve obtained from previous experiments (Refs. 1–3) with present work added.

same as the Long and Meyer transitions, but there are differences to be kept in mind. Long and Meyer reported transitions only for $P/P_0$ > 0.15, while we find onset $A$ continuing down to $P/P_0 \simeq 0.02$. Long and Meyer used flow channels of stainless steel and platinum in glass: These surfaces were probably not more uniform than conventional substrates.

We have translated our $P/P_0$ values to film thicknesses according to a $He^4$ vapor-pressure isotherm at 2 K taken in a different Grafoil cell.[17] Those results, combined with directly measured layer capacities, are well described by a Frenkel-Halsey-Hill isotherm[18] $P/P_0 = \exp(-\alpha/n^3)$ over the full experimental range up to $n \simeq 10$ layers. The empirical $\alpha = 67.5$ is consistent with the empirical binding of $He^4$ on graphite. According to this calibration onset $A$ occurs for films of about 2.5 layers or more. The thicknesses at which onset $B$ is seen lie in the range from about 4 to 7 layers; for example, at $n = 5.1$ layers, $T_B = 1.86$ K.

The occurrence of two distinct flow transitions is novel, although liquid $He^4$ in porous Vycor shows a superfluid transition at $T < T_\lambda$ and yet has a density maximum at $T_\lambda$.[19] The precursor has not been previously seen in He film flow, but it seems analogous to the rise in electrical conductivity at $T > T_c$ of thin-film superconductors[20-22] ascribed to the greater role of fluctuations in lower-dimensional systems.[23]

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