Vortex Core Size in Submonolayer Superfluid ^{4}He Films

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The superfluid density of ^{4}He films adsorbed on 500 Å alumina powder is measured for films with transition temperatures between 50 and 700 mK. The transitions are controlled by the universal Kosterlitz-Thouless line, but a strong increase in the broadening of the transition is observed for the thinnest films. Analysis shows that this broadening results from a vortex core size that increases rapidly as the film is thinned, scaling roughly with the interparticle spacing of the superfluid submonolayer. This system is evidence of a three-dimensional superfluid transition mediated by vortex excitations.

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It is now well established that the superfluid phase transition of thin helium films adsorbed on two-dimensional flat substrates is caused by the thermal excitation of vortex pairs, as proposed by Kosterlitz and Thouless (KT) [1]. The vortices drive the superfluid areal density σ_s abruptly to zero at a critical temperature T_{KT} when the quotient $\sigma_s\,(T)/T$ approaches the universal value

$$(\sigma_s/T)_{\mathrm{KT}} = 2m^2 k_B/\pi\hbar^2 = 3.49 \times 10^{-9}\ \mathrm{g/cm^2\,K}, \quad (1)$$

where m is the ^{4}He atomic mass. The sudden drop of the superfluid density to zero at precisely this value has been confirmed in both third sound and torsion oscillator measurements [2]. These experiments find a linear increase in T_{KT} with the coverage of added helium, which agrees with Eq. (1) since σ_s is the mass per unit area of the superfluid portion of the film.

However, the nature of the superfluid transition of helium films adsorbed in porous materials has been a subject of considerable debate. In such a geometry the film becomes multiply connected and three dimensional on length scales larger than the grain size of the substrate. Initial torsion oscillator measurements [3] at Cornell University with films adsorbed in porous Vycor glass found behavior different from that of flat films: The superfluid density did not fall abruptly to zero at the transition, but decreased smoothly, varying as a power law of the temperature difference from the critical point, similar to the bulk ^{4}He superfluid transition. In the thinnest films an additional broadening of the transition was noted [4]. The hypothesis was advanced that this additional broadening signaled a crossover to an ideal dilute Bose gas as the particle density in the superfluid portion of the film was reduced, and theoretical support for this idea was formulated [5].

More recent experiments [6–9] and theories [10–12] have focused instead on an alternative hypothesis that in a porous material the Kosterlitz-Thouless vortex-pair mechanism is altered by the porous geometry, with the result that the sharp jump in σ_s is smoothed out. Evidence for the KT mechanism came from third sound measurements [6] on films adsorbed on alumina powders, which showed that the phase transition begins at the universal KT line given by Eq. (1), but with a smooth decrease of the superfluid density to zero at a critical temperature T_c higher than T_{KT}. It was also shown that the magnitude of $T_c - T_{\mathrm{KT}}$ varied with the grain size of the alumina powder, decreasing considerably as the grain size increased, approaching the sharp KT jump in the two-dimensional limit of infinite grain size [8]. Later measurements [9] found that the same behavior occurs in porous glass substrates when the pore size is varied. Neither the correspondence with the KT line nor the variation with grain size can be accounted for by the Bose gas hypothesis [13] of Refs. [4] and [5], whereas these are primary predictions of the vortex models [6,10–12].

In an attempt to understand the additional broadening reported [4] in the thinnest films on the Vycor substrate, we have undertaken detailed measurements of the areal superfluid density of a series of films adsorbed on 500 Å diameter alumina powder. The transition temperatures of these films ranged from 50 to 700 mK, a regime where the superfluid thickness of the film is less than a monolayer. As the film thickness is reduced, we find a continuous broadening of the transition (i.e., an increase of $T_c - T_{\mathrm{KT}}$) that becomes most apparent in the thinnest films. An analysis using the KT theory modified for porous materials shows that this broadening is readily explained in terms of a vortex core size that increases rapidly as the film is thinned, in agreement with an earlier prediction [14]. A brief description of our initial results has been published elsewhere [15].

The measurements are carried out by monitoring the shift in the period of a torsion oscillator as the superfluid portion decouples from the oscillating mass, the technique developed in Refs. [3] and [4]. The Al_2O_3 powder, Linde 0.05 C, is the same powder of nominal diameter 500 Å used in the third sound measurements [6]. It is formed into a solid plug using a slip-casting technique described elsewhere [16] and is then machined into a cylindrical shape of diameter 1.26 cm and length 1.28 cm. The slip-cast sample is more densely packed than the previous pressure-packed powders, with a porosity of 59%, and we believe that it is more homogeneous [15]. The surface area of the sample is 146 m^2, derived from

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the nitrogen isotherm measurements of Ref. [17]. The sample is secured inside a thin-walled aluminum can, and the can is epoxied to a beryllium copper torsion bar 2 mm in diameter. The torsion mode at 1.50 kHz is driven and detected capacitively, with a phase-locked loop maintaining a constant oscillator amplitude, and the period is measured using an oven-controlled frequency counter. The Q factor of the oscillator is about 1.5×10^6 under operating conditions, and the resolution of the period measurement is about 2–3 ps. The cell temperature is swept under computer control, with temperature step sizes in the range 0.1–1.0 mK, and a minimum equilibration time of 40 min is allowed after each step.

Figure 1 shows the results for the measured period shifts ΔP as a function of temperature for a number of films with helium coverages ranging from 35.6 to 45.2 μmole/m^2. The first 34.8 μmole/m^2 of coverage is the nonsuperfluid "inert" layer, and superfluid period shifts are detected only for coverages above that value. By extrapolating these period shifts to $T = 0$ the sensitivity of the oscillator to added superfluid mass is found to be $\Delta P/\sigma_s = 12.5$ ns/(ng/cm^2). This calibration of the data in terms of areal superfluid density is shown on the right axis of Fig. 1. The solid line in the figure is the KT critical line of Eq. (1); these data corroborate the earlier third sound results [6] that the transition begins at the critical KT point, but falls to zero smoothly at a T_c shigher than T_{KT}. We observe a rounding of the transtion near T_c of order 10–15 mK, which is larger than that observed in the Vycor studies [3]. This rounding may be the result of remaining density inhomogeneities across the sample.

The curves in Fig. 1 are not self-similar, with the lowest-coverage films being discernibly more broadened out than those with higher transition temperatures. This is better illustrated in Fig. 2, in which the data have been scaled by the values ΔP_{KT} and T_{KT} where the KT line crosses the curve for each film thickness. In the region of reduced temperatures greater than 1.0 a continuous broadening of the transition can be seen for all of the films as the coverage is reduced, increasing rapidly for the thinnest films with the lowest values of T_{KT}. In the low-temperature regime of reduced temperatures less than 1.0 there is some variation of the reduced period shifts for the thicker films, but for films with T_{KT} less than 250 mK the low-temperature data appear to be converging on a single curve.

To analyze the broadening of the transition seen in Fig. 2 we employ the KT vortex theories modified for the case of porous geometry [6,10–12]. The main point is that a new length scale enters the problem, the average radius R of the powder grains. Vortex pairs of separation less than the grain size are essentially unchanged from the flat-substrate case, having an excitation energy logarithmic in their separation. Pairs with separation larger than the grain size, however, are substantially modified, with an excitation energy increasing linearly with separation [10,11]. Such pairs, dubbed "strings," can be oriented three dimensionally, since they reside on different grains, but they are still dipole objects that act to reduce the superfluid density [12]. Since the strings are energetically costly, they act to effectively cut off the KT renormalization equations at the scale of the grain size R, where the crossover from pairs to strings occurs. This leads to a finite-size broadening of the transition, with the degree of broadening depending on the parameter R/a_0 where a_0 is the vortex core radius. This scaling was first pointed out in Ref. [6], where a model of the KT transition on a single sphere of radius R was used to account for the broadening observed in the third sound measurements, and values of a_0 were extracted from fits to the data. The addition of string excitations to this model retains the finite-size broadening effect, but with the change that a fully three-dimensional phase transition takes place. In the region near T_c strings can finally be thermally excited, and they unbind and drive the superfluid density to zero [12]. Although a linear potential is normally confining, the strings can unbind because the screening of large strings by smaller ones reduces the effective potential to a variation that is slower than linear [18].

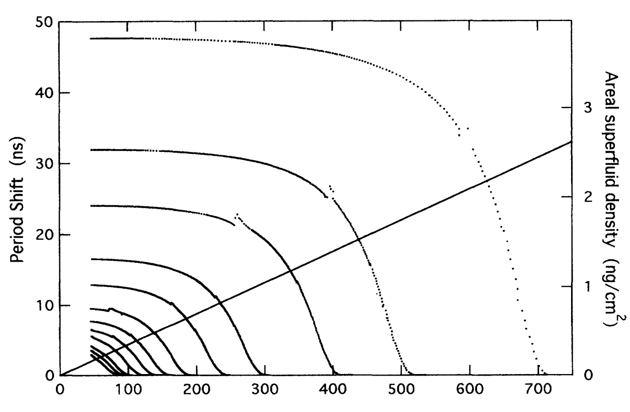

FIG. 1. Measured period shifts (left axis) of the torsion oscillator as a function of temperature for a number of different film coverages. The right axis shows the calibration in terms of the areal superfluid density.

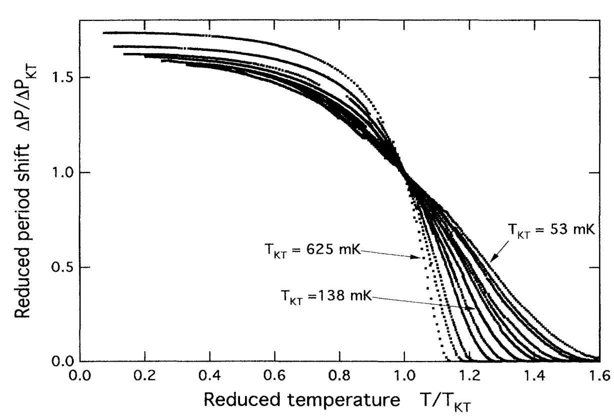

FIG. 2. The data of Fig. 1 scaled by the values ΔP_{KT} and T_{KT} where the curves cross the KT line.

Fits of the data of Figs. 1 and 2 have been carried out using the single-sphere model of Ref. [6] supplemented by the string model of Ref. [12]; an example of those fits is shown in Fig. 3 for the film with $T_{KT} = 235$ mK. Four parameters are involved in the fits: the zero-temperature intercept $\sigma_s(0)$, the "bare" superfluid density $\sigma_s^0(T)$, the core parameter a_0, and the core energy E_c. The first two parameters are fit separately in the low-temperature regime $T/T_{KT} < 0.5$, where no vortices are excited. The variation of σ_s^0 is well described by $\sigma_s^0 = \sigma_s(0)[1 - \beta(T/T_{KT})^2]$, as also found in the Vycor measurements [3], with β varying from 0.065 in the thickest film to 0.142 for the films with T_{KT} less than 250 mK. In conjunction with least-squares fitting, the single-sphere model is then used to find the best-fit values of a_0 and E_c in the high-temperature regime, fitting only to data points with a reduced period shift greater than 0.4 to avoid the finite-size "tail" region of the model. The core energy is primarily determined from the points with reduced temperature near 1.0, while a_0 is determined by the degree of broadening in the decrease of σ_s at higher temperatures. The best-fit values for the data set of Fig. 3 are $a_0 = 82$ Å and $E_c/K_0 = 2.35$, where $K_0 = \hbar^2\sigma_s^0/m^2 k_B T$. Also shown in Fig. 3 is the superfluid density calculated using the string model of Ref. [12], but with the difference that the single-sphere recursion relations are used to the crossover point (where the pairs are a sphere diameter apart), and then the string recursion relations are iterated to larger length scales. This calculation gives a power-law variation of σ_s near the transition, with a superfluid exponent of 0.64, close to the presumably more accurate value 0.67 predicted in Ref. [11]. As can be seen in the figure, the rounding of the data prevents us from checking this exponent.

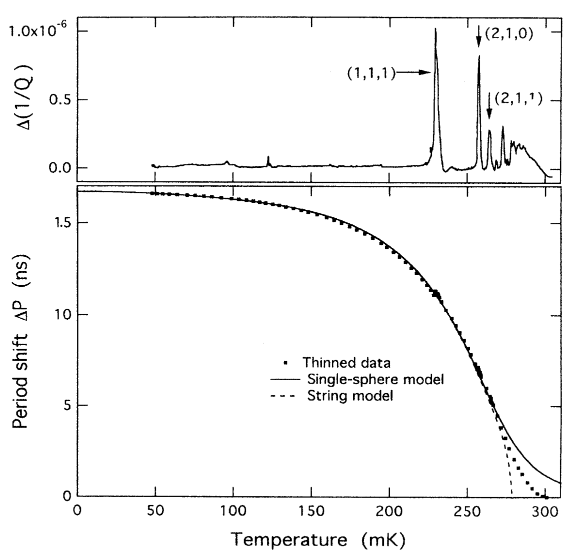

FIG. 3. Thinned data (about 20% of the total) of the period shift and change in inverse Q for the film with $T_{KT} = 235$ mK.

The small perturbations in the data of Figs. 1 and 3 (and edited out of Fig. 2) are third sound resonances crossing the torsion mode frequency. The mode-pulling effect on the torsion period is a source of uncertainty in performing the data fits, and the points around the perturbations are given less weight in the fits. The third sound modes are even more evident as sharp changes in the inverse Q of the oscillator (monitored by the drive voltage needed to keep the amplitude constant), shown in the upper graph of Fig. 3. These resonances are readily identified as those with mode numbers (m, n, n_z) of the cylindrical sample, characterized by the nth node of the derivative of the Bessel function J_m. Modes with $m = 0$ are not observed, as expected, since they are purely radial and should not couple to the torsional mode. In the temperature region where σ_s falls to zero there is an additional broad attenuation peak in the $1/Q$ data, upon which the third sound modes of rapidly decreasing amplitude are superimposed. This peak can be identified in all of the data curves, and we believe that it arises from the finite-frequency dissipation of the vortex pairs, as predicted theoretically [6,19]. A similar attenuation peak was observed on porous glasses [7,9].

Figure 4 shows the results of the data fits for the parameters a_0 and E_c/K_0 plotted versus T_{KT}. Since T_{KT} is proportional to the superfluid coverage this plot should be viewed as primarily the variation of a_0 and E_c with coverage, although, in principle, there could also be some temperature dependence of these quantities. The agreement with the previous third sound results [6] in thicker films is quite reasonable, considering the difference in measurement techniques. The rise in the core energy as T_{KT} decreases from 620 to 235 mK is well resolved and is responsible for the low-temperature variation between the data sets seen in Fig. 2. Much more uncertain is the subsequent decrease of E_c at lower coverages, due to difficulties in extrapolating the lowest curves to $T = 0$ as needed for the fitting procedure. An extension of the measurements to lower temperatures will be necessary to decide if this downturn in E_c is real.

The rapid rise in a_0 seen in Fig. 4 as the coverage is reduced is the source of the continuous broadening of the transition in Fig. 2. This rise in a_0 was anticipated [14] from the observation that the superfluid thickness of these films is less than a monolayer for coverages with $T_{KT} <$ 0.8 K, and therefore reduction of the coverage increases the average spacing between atoms in the superfluid layer. The vortex core is a node of the macroscopic superfluid wave function; from general properties of the many-body wave equation it is well known that the ground-state wave function cannot vary more rapidly than the interparticle spacing. Solutions of the Gross-Pitaevskii equation for the vortex structure of Bose particles with a delta-function interaction [20] give a core size increasing linearly with the interparticle spacing r_0 of the submonolayer, varying as $a_0 = r_0(\hbar^2 d/2mV_0)^{1/2}$, where V_0 is the strength of the potential and $d \approx 3$ Å is the atom size. The spacing

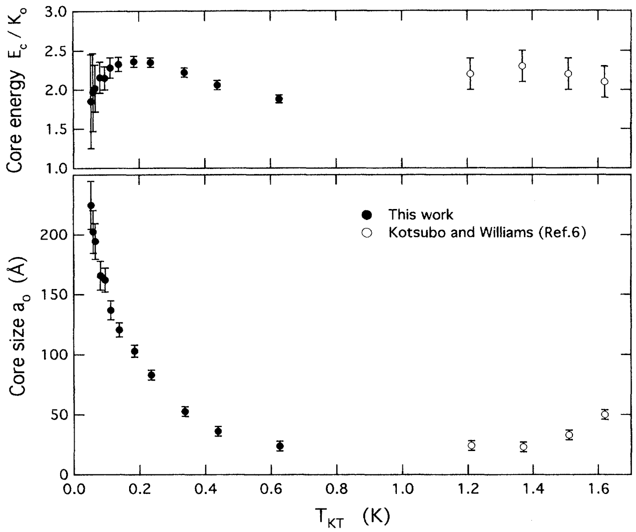

FIG. 4. Variation of the vortex core size and core energy with the superfluid coverage, which is proportional to T_{KT}.

r_0 varies as $(m/\sigma_s)^{1/2}$, and from Eq. (1) this yields a core size that should increase as $(T_{\mathrm{KT}})^{-1/2}$, if there is no other intrinsic temperature variation of a_0. Fitting the core-size data of Fig. 4 by the form $(T_{\mathrm{KT}})^{-\alpha}$ yields an exponent $\alpha = 0.65 \pm 0.1$, at least in rough agreement with the expectation that it should scale as the interparticle spacing. There have been other experimental observations of an increasing vortex core size that corroborate the data presented here. The unpublished data of McQueeny [21] taken with films adsorbed on Millipore filter paper (with cylindrical fibers of diameter 500–2000 Å) was analyzed using the theory of Ref. [11], with results for the vortex core size that are very similar in both magnitude and temperature dependence to our results in Fig. 4. An increasing core size was also deduced in Ref. [14] from fits to the Vycor data.

In the fits by the single-sphere model, it is assumed, for lack of better knowledge, that the pair-to-string crossover distance is the average grain diameter of 500 Å. The actual porous geometry, however, is not well modeled by a single sphere. At the packing density of our sample there are on the average 4–5 powder grains touching a given grain, and the distance a pair could separate before exciting appreciable flow on a neighboring grain might well be more on the order of $\frac{1}{5}$ of the sphere diameter. Since the broadening in the model is a function of the ratio of the crossover length to the core size, a decrease of the crossover length would decrease the fitted core size by the same factor. Dividing the core sizes of Fig. 4 by the above factor of 5 would bring their magnitude to within a factor of 2 of the interparticle spacing r_0. We think that this could be a plausible scenario, but detailed simulations of the vortex hydrodynamics in a more realistic model of the porous geometry will be needed to decide the actual crossover length that should be used.

In summary, we have observed a continuous broadening of the superfluid transition of helium films as the coverage is reduced. The correlation with the universal KT line shows that a vortex mechanism is involved, and the data are well explained by a crossover from vortex pairs to three-dimensional vortex strings at the powder grain size. The broadening is the consequence of an increase in the vortex core size as the interparticle spacing in the submonolayer superfluid is increased. Since the Cornell Vycor data have also been shown to correspond with the KT critical line [6,21,22], we speculate that the additional broadening observed in those data arises from the same mechanism, an increase in the vortex core size. The present results provide experimental evidence that the superfluid transition in porous materials involves three-dimensional vorticity, a finding that has implications for the nature of the superfluid λ transition in bulk helium [18].

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