Superfluid transition of ⁴He films adsorbed on multiply connected surfaces

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A Kosterlitz-Thouless-like approach is used to describe the superfluid transition of thin ⁴He films adsorbed on multiply connected surfaces such as packed powders or other porous materials. The elementary topological defect mediating the transition is a string linking two vortices of opposite circulation. The long-range interaction between strings gives rise to a phase transition which has a critical exponent for the superfluid density of v=0.64.

Kosterlitz and Thouless¹ (KT) first demonstrated the key role played by vortices as the elementary topological excitations in the superfluid phase transition of ⁴He films. In their picture the transition temperature $T_{\rm KT}$ separates a low-temperature superfluid state, where vortices of opposite circulation are bound in pairs, from a hightemperature normal state where single free vortices exist. For a flat two-dimensional (2D) film, this theory predicts² that the areal superfluid density σ_s jumps at $T_{\rm KT}$ from zero to a finite universal value $\sigma_s = (2/\pi) (m_4/\hbar)^2 k_B T_{\rm KT}$. Below $T_{\rm KT}$ the density is sensitive to the interaction between pairs, and a real space renormalization procedure³ allows a calculation of the macroscopic, observable σ_s^r , starting from the "bare" microscopic value σ_s^0 at the shortest length scales. The predictions of the theory have been well verified by experiments on flat substrates of centimeter dimensions.

However, notable differences with the pure 2D case have been found on substrates characterized by a small length scale such as packed powders 5^{-7} or other porous materials.⁸⁻⁹ The onset of superfluidity occurs at a temperature close to the KT prediction, but the superfluid jump is broadened and the density decreases continuously to zero over a temperature domain of width ΔT . In order to interpret their data in Vycor glass, Reppy and coworkers⁹ proposed that the transition becomes similar to the bulk three-dimensional (3D) λ transition, crossing over to a dilute Bose gas transition in the thinnest films. However, the theory based on these ideas¹⁰ does not explain the observed decrease of ΔT when the substrate grain size is increased, 5-7 and, thus, cannot be extrapolated to the flat-substrate KT case which must be recovered in the limit of very large grains.

On the other hand, Kotsubo and Williams⁵ were able to account for the broadened transitions they observed in packed powders by employing a finite-size modification of the KT theory, in which only vortex pairs of separation smaller than the grain size are included in the renormalization. They found a transition width ΔT in agreement with the prediction of Barber,¹¹

$$\Delta T = \left(\frac{B}{\ln(a/a_0)}\right)^2,\tag{1}$$

where a is the grain size, a_0 the vortex core size, and B the nonuniversal constant of the Kosterlitz correlation length. In the limit $a \rightarrow \infty$ the sharp flat-substrate transition is recovered. The defect of this model is that it does not take into account the possibility of vortex pairs with separation greater than the grain size. As a result, the transition predicted by the model is not a full thermodynamic transition, displaying a finite-size "tail" rather than power-law behavior in the critical region.

In the present paper we present a model of the superfluid transition which accounts for the 3D connectivity of the surface. Following the work of Minoguchi and Nagaoka¹² and Machta and Guyer,¹³ we introduce a vortex string as the elementary excitation of the system. The string connects two 2D vortices of opposite sign through a single path along the substrate. However, unlike the purely 1D strings of Minoguchi and Nagaoka, our strings interact with each other through a long-range dipole flow field. This leads to a 3D thermodynamic phase transition, while the 1D case is rigorously known not to have a phase transition.¹⁴

We model the porous medium as a 3D lattice made up of intersecting cylinders, as shown in Fig. 1. For simplicity, we assume in the following that there is no distribution in the cylinder sizes; they all have radius a and length bbetween intersections (a - b). The interaction energy Ubetween a vortex and an antivortex in the helium film coating this surface takes different forms depending on



FIG. 1. Lattice model of a porous medium, made of intersecting cylinders (here a cubic arrangement for example). The circulation must be quantized around the closed paths C1 or C2.

On the other hand, when the vortex separation is larger than the lattice spacing $(R \gg a, b)$, the flow pattern has to account for the nontrivial connectivity of the surface. At the same time, the circulation of the flow around any closed path on the surface (like C1 or C2 of Fig. 1) has to be quantized. We believe that it is possible to satisfy these conditions by drawing a closed line forming a loop in the 3D space [labeled C in Fig. 2(a)], which crosses the film only on both cores of the vortices, and such that the circulation around any closed path on the surface is one if it passes through this loop, and zero otherwise.

In the following, we neglect fluctuations and limit ourselves to the lowest energy excitations, as shown in Fig. 2. The ring joining the 2D vortices separated by R must then have a minimum area, and is made of two straight and parallel strands, separated by their minimum distance $\sim b$ [Fig. 2(b)]. One of the strands corresponds to quantized circulation around cylinders, and the other one to opposite circulation around loops, together forming a "string" joining the 2D vortices [Fig. 2(a)]. At large distance $r \gg b$ from this string, the flow field is the same as the one generated by a long and thin dipolar ring, and decays like $1/r^2$. Besides the core energy of the 2D vortices, the total energy U of this excitation is equal to the total kinetic energy associated with the superfluid flow. It can be roughly divided in two parts: one comes from the flow in the cen-



FIG. 2. (a) String structure of lowest energy, involving a +1 quantum around a cylindrical trunk, and -1 around one adjoining loop. The circulation about any closed path circling the dashed loop (C) is different from zero. For simplicity the far-field dipolar flow is not shown here. (b) In the continuum limit this structure is equivalent to a long thin ring stretched between the two vortices.

tral elements of the string, the other one from the far field dipolar flow. Although an exact computation of these two parts can only be achieved numerically, it is easy to see that they are both proportional to the vortex separation Rand to the reduced superfluid density $K = (\sigma_s/k_BT)$ $\times (\hbar/m_4)^2$, so that we simply write $U = \alpha \pi K R/b$. The dimensionless factor α only depends on the geometry of the lattice, and increases with the connectivity of the substrate. It can be shown that $\alpha > b/a > 2$.

We use a Kosterlitz-Thouless approach to calculate the macroscopic superfluid density σ'_s , renormalized by the interactions between strings. We consider only straight strings, neglecting their fluctuations. A magnetic analogy¹⁵ is used to compute the interaction energy ΔU between a fixed long string and a string of shorter length R which experiences the flow field v_s generated by the long string

$$\Delta U = -\frac{\rho_s}{k_B T} \mathbf{m} \cdot \mathbf{v}_s \,. \tag{2}$$

Here, $\mathbf{m} = 2\pi(\hbar/m_4)\mathbf{S}$ is the dipole moment of the ring formed by the two strands of the string, with $\mathbf{S} = bR\hat{\mathbf{n}}$, and $\rho_s \sim \sigma_s/b$ is the average 3D superfluid density per unit volume of the sample. Taking a thermal average over the possible orientations of **m** gives

$$\langle \Delta U \rangle = -\frac{1}{3} \left[\rho_s \frac{m v_s}{k_B T} \right]^2. \tag{3}$$

The probability dp to find a current loop of length between R and R + dR at a given position within the volume $d^{3}r$ is

$$dp = A \exp(-\alpha \pi K R/b) \frac{4\pi R^2 dR}{b^3} \frac{d^3 \mathbf{r}}{b^3}.$$
 (4)

When R = b this reduces to the probability of finding a pair of vortices separated by a cylinder length, which determines the normalization constant as $A = \exp(-2E_c + \alpha \pi K)$. E_c is the vortex core energy already renormalized up to the scale b, and may be computed from the bare core energy E_c^0 on the atomic scale using the recursion relations for vortices on a single cylinder.^{5,12,13} We introduce for convenience the usual vortex fugacity $y = \exp(-E_c)$.

The renormalized energy of the fixed string is $U_r = U - \int \langle \Delta U \rangle dp$. Defining $K_r = (\hbar/m_4)^2 (\sigma_s^r/k_B T)$, using the dipole flow field $v_s = (\hbar/m)(b/r^2)$, and integrating over $d^3\mathbf{r}$ gives the renormalized superfluid density,

$$K_r = K - \beta \int_b^\infty K^2 y^2 \exp[-\alpha \pi K (R/b - 1)] \frac{R^4 dR}{b^5}.$$
 (5)

Here the constant β is of the order of $16\pi^3/3$, within a factor of order unity depending on the particular lattice structure. For self-consistency the K's in the integral should be replaced by the renormalized value K_r . Expanding in powers of y^2 , Eq. (5) is then the first two terms of the expansion. Near the phase transition where $K_r \rightarrow 0$ the integral becomes large and the expansion breaks down. We use the technique of Jose, Kadanoff, Kirkpatrick, and Nelson³ in this regime, dividing the integral into two parts, one going from b to $b' = be^{\delta}(\delta \ll 1)$, and the other from b' to ∞ ,

$$K_{r} = K - \beta K^{2} y^{2} \delta - \beta \int_{b'}^{\infty} K^{2} y^{2} \exp\{5\delta - \alpha \pi K [(R - b'e^{-\delta})/b'e^{-\delta}]\} \frac{R^{4} dR}{b'^{5}}.$$
(6)

This takes the same form as Eq. (5) under the transformation

$$K \rightarrow K' = (K - \beta K^2 y^2 \delta) e^{\delta},$$

$$y^2 \rightarrow y'^2 = y^2 e^{(4 - a\pi K)\delta},$$

$$K_r \rightarrow K'_r = K_r e^{\delta},$$
(7)

Repeating this transformation indefinitely, it can be written in differential form, setting $\delta = dl$,

$$\frac{\partial K}{\partial l} = K - \beta K^2 y^2,$$

$$\frac{\partial y^2}{\partial l} = (4 - \alpha \pi K) y^2,$$
(8)

and the observable superfluid density is

$$K_r(l=0) = K_r(l)e^{-l} = \lim_{l \to \infty} K(l)e^{-l}.$$
 (9)

This last equation is the Josephson hyperscaling relation in three dimensions,² meaning that this phase transition will be accompanied by a critical specific heat peak.

Equations (8) have a trivial fixed point K = 0, y = 0, and a nontrivial one $K_c = 4/\pi \alpha$, $y_c^2 = 1/\beta K_c$. Depending on the initial value K_0 at the atomic scale, the trajectories near the fixed point either lead to $K \rightarrow \infty$, $y \rightarrow 0$ (finite superfluid density, vortices bound on stings) or to $K \rightarrow 0$, $y \rightarrow \infty$ (zero superfluid density, vortices unbound with strings of infinite length). At a critical value K_{0c} corresponding to the transition temperature T_c the trajectory ends on the fixed point (K_c, y_c) . The superfluid density goes continuously to zero as T approaches T_c , and by linearizing the recursion relations about the fixed point one finds $\sigma_s \sim (T - T_c)^v$ with $v = 2/(\sqrt{17} - 1) = 0.64$. We show in Fig. 3 the variation of $\sigma_s(T)$ for the case most similar to a packed powder geometry, b=2a. For scales less than 2a the usual KT recursion relations are iterated,¹⁶ and then at longer scales Eqs. (8) are iterated to macroscopic lengths. The different curves correspond to different values of $2a/a_0$, the ratio of the grain diameter to the vortex core parameter. We used the particular values of the parameters $\alpha = 2$, $\beta = 16\pi^3/3$ and $E_c^0 = 2.2K_0$. We have also noticed that the transition temperature is somewhat affected by the value of α , i.e., the connectivity of the lattice. This result could be experimentally checked by varying the packing coefficient of the powders. The broadening $\Delta T \sim T_c - T_{\rm KT}$ from the pure KT transition agrees with Eq. (1) and results from the effective cutoff of the recursion relations at the grain size, as discussed in Refs. 5, 12, and 13. The cutoff results in this case from the changeover to the more energetically costly strings. However, as $\sigma'_s \rightarrow 0$, the strings finally lead to a full thermodynamic phase transition at T_c .

This monotonic broadening of the KT transition with grain size has been observed in all porous materials studied to date, 5^{-9} ranging from 50- μ m Al₂O₃ powder to 60-Å Pt powder. We note that the predicted exponent

calculation is in good agreement with th found by Reppy and co-workers^{8,9} i ass of
$$\sim 150$$
-Å grain size. Higher value

v = 0.64 of our of e result v = 0.63n porous Vycor gl of the exponent (up to one) have been observed in packed powders,⁶ and we do not know how the distribution of grain sizes in these systems might affect the exponent, a factor which has not been included in our model. A more complete theoretical treatment should also take into account the fluctuations of the strings, using a configurational sum over the different possible paths. Some initial calculations along this line suggest that the average fluctuation amplitude remains small even near T_c , but we do not know the extent that v will be affected.

It is possible that our model is related to the Villain model formulation of Machta and Guyer,¹³ although they are not exactly equivalent. Indeed, our string is a superposition of azimuthal and axial pore vortices, which are their elementary excitations. However, their calculation only takes into account the axial pore vortices, while we find it necessary to include both kinds of pore vortices in constructing strings having 2D vortices at the end points. In addition, we feel that our model has the advantage of giving a clear physical picture of the transition. For example, the diverging correlation length near T_c can easily be identified as the mean length of the strings being excited, which becomes infinite at T_c .

In summary, we have shown that the superfluid transition of ⁴He films adsorbed on multiply-connected surfaces can be simply described in a vortex pair formulation. The vortices are linked by a string along the substrate, and are the excitations responsible for the thermodynamic phase transition. The predictions derived from the recursion relations for the superfluid density are in agreement with present experimental data.



FIG. 3. The renormalized superfluid density plotted vs reduced temperature, resulting from iterating the KT equations to scale b = 2a, and then Eqs. (8) to infinite scale. The curves a, b, and c correspond to increasing grain size $(2a/a_0=6, 30, 150, re$ spectively), and curve d to the pure KT case $(2a/a_0 \rightarrow \infty)$.

4675

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